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ABSTRACT

An essentially self-contained treatment of the theory of low-energy neutron scattering is given with emphasis on the extent to which symmetries and dynamics of the scattering system may influence neutron-nuclear collision processes. The general problem is formulated using the "Born Approximation" and Fermi pseudo-potential, and calculations are made for two crystal models. The simpler Einstein crystal is found to be adequate in qualitatively explaining the measured scattering cross section. For problems sensitive to dynamical features of the scatterer, a more realistic crystal model is carefully derived, and the associated problem of normal mode analysis, with both standing and traveling waves, is discussed in detail. It is shown that the more elaborate model gives results useful in the interpretation of the experimental studies of lattice dynamics, particularly in the direct determination of dispersion relations and frequency distributions. In obtaining the various cross sections the concept of coherent scattering in contrast to that of interference scattering is clearly illustrated; suitable approximations and meaningful limiting cases are obtained. Incoherence due to isotopic mixture and nuclear spin is also discussed. It is shown that the results of a semi-classical treatment are readily interpretable and should be helpful in a general understanding of the interaction of low-energy neutrons with solids.

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I. INTRODUCTION

In recent years considerable attention has been directed towards the development of neutron diffraction as a solid-state physics research tool. As with other techniques, such as ultrasonics, optical measurements, electron- and nuclear-spin resonances, and Mossbauer effect, valuable information about the scattering system can be obtained from a neutron diffraction experiment. For example, one of the most significant studies made with neutrons thus far is the direct determination of dispersion relations and frequency distributions for the normal modes of the crystal. The usefulness of these measurements, as is the case in general, necessarily depends upon a suitable theoretical model for interpretation.

Excellent fundamental(1) and review(2) papers on the theory of slow-neutron scattering by crystals are available in the literature; nevertheless, it seems to us that a discussion of some of the fundamental aspects is lacking. Consequently we attempt to present in this paper an essentially self-contained treatment of neutron interaction in an acoustic field, the main purpose being to illustrate the extent to which the scattering is influenced by symmetries and dynamics of the scattering system. It will be seen that the present problem contains many of the general features of interaction of radiation with matter. For example, it provides a systematic comparison with the related problem of electron interaction in an electromagnetic field. Therefore it may be possible that, when viewed from the proper prospective, neutron diffraction studies can be related to those using the other above-mentioned techniques. We also hope that the following work will enhance the usefulness of the existing literature.

In Section II the differential scattering cross section is obtained by considering, somewhat classically, the scattered neutron waves from each scattering center in the crystal. Then after the general problem of neutron scattering by systems is formulated in Section III the quantum results based on the Einstein crystal model are discussed in the next section. A considerably more realistic dynamical model for crystals is developed in Section V, followed by a section on normal coordinate transformation in which the decomposition of particle coordinates in terms of annihilation and creation operators is explicitly derived. A detailed calculation of the various elastic and inelastic cross sections for a monatomic crystal containing spinless particles is given in Section VII where some limiting cases will be obtained. The final section contains a discussion of spin effects.

II. A SEMI-CLASSICAL APPROACH

A detailed analysis of neutron scattering by macroscopic systems is necessarily quite complicated. It is therefore instructive to present a preliminary discussion in which rigor and a certain amount of details can be sacrificed for physical insight. In this section we will concern ourselves with such an examination of low-energy neutron scattering by crystals. The treatment is admittedly of an intuitive and semi-classical nature; nevertheless, meaningful results can be obtained. Since the calculation and results for a more elaborately formulated problem will be discussed later, we will not dwell on details of interpretation beyond what is sufficient for illustrative purposes.

In order to develop a theory of neutron scattering it is essential to **first** determine in what way are the calculations to be ultimately correlated with measurements. Clearly, before we can begin with any theoretical consideration we should know what type of experiment must the theory explain. Let us therefore consider the following idealized experiment. A beam of monoenergetic neutrons impinges upon a scattering system; the interaction between the neutrons and target results in some neutrons being deflected from the original path with energies greater or less than that before the collision. The scattered neutrons are detected by a counter (see Figure 1). If the counter is energy-sensitive, then the scattered neutrons with a particular energy may be detected, and in this manner an energy distribution can be obtained. To avoid unnecessary complications we will assume that the target-detector separation is large compared to all linear dimensions of the scattering system and that multiple collisions can be ignored.

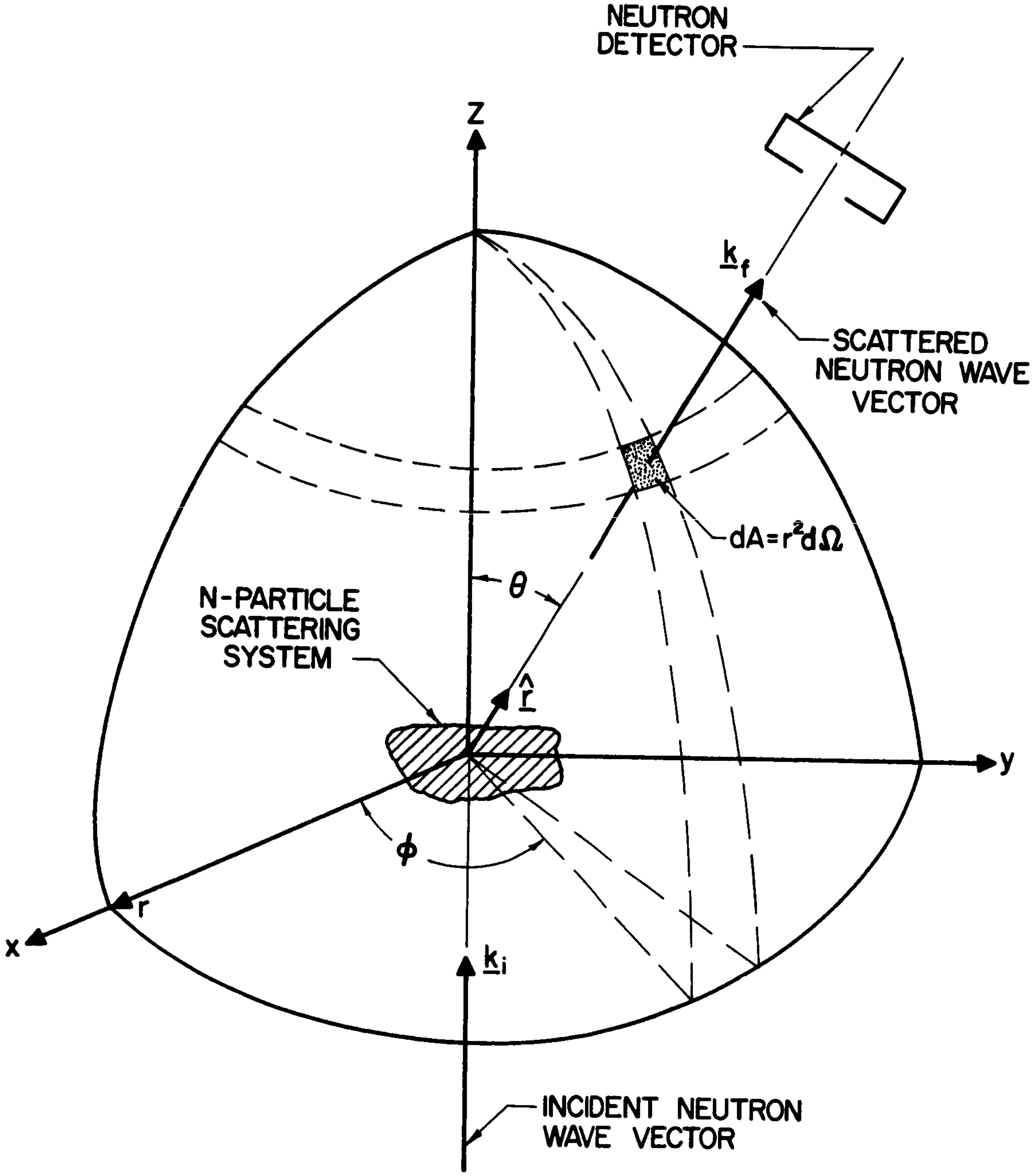

Figure 1. Schematic Diagram of Neutron Scattering Experiment.

It is obvious that a fundamental quantity which characterizes such a measurement is the probability that an incident neutron with given initial energy will be scattered into a given direction. That is to say, we can describe the scattering experiment in terms of a differential cross section $\sigma d\Omega$, defined as the number of neutrons per second scattered into an element of solid angle $d\Omega$ about direction $\hat{\underset{\sim}{r}}$ divided by the product of number of incident neutrons per cm^2 per second and total number of scatterers. By this definition $\sigma d\Omega$ depends upon the initial neutron energy and the angle of scattering. Later we will want to describe measurements using energy-sensitive detectors and will then introduce the differential cross section which also depends upon final neutron energy.

Since the radiation to be scattered and measured consists of a beam of neutrons, then what we wish to calculate is the scattered current, given by the scattered beam intensity multiplied by the group velocity of the traveling waves. If we represent the incident and scattered beams as traveling waves ψ_i and ψ we obtain

$$\sigma d\Omega = \frac{r^2 \ |\psi|^2 \ \hat{\underset{\sim}{r}} \cdot \underset{\sim}{k}_f}{N \ |\psi_i|^2 \ k_i} \, d\Omega \ , \tag{II.1}$$

where $\underset{\sim}{k}_i$ and $\underset{\sim}{k}_f$ are incident and scattered wave vectors and N is the number of scatterers. It is seen that given an incident wave the problem is to determine the scattered wave and to exhibit its dependence upon the geometric and dynamical features of the scattering system.

Consider a system of particles (nuclei) in which the position of the ℓ-th particle is specified by $\underset{\sim}{R}_\ell$. Let the incident neutrons be represented by a plane wave $Ae^{i(\underset{\sim}{k}_i \cdot \underset{\sim}{r} - \omega_i t)}$ with amplitude A, propagation vector

$\underset{\sim}{k}_i$ and circular frequency ω_i. For convenience we will put the origin of our coordinates at one of the particles so that the incident wave at this particle is just $Ae^{-i\omega_i t}$ (see Figure 2). The incident wave at the ℓ-th particle is then $Ae^{i(\underset{\sim}{k}_i \cdot \underset{\sim}{R}_\ell - \omega_i t)}$. It is known that neutron-nuclear scattering is essentially a hard-sphere interaction, and this implies that at the surface of the nucleus the incident and scattered waves are the same. In other words, the phase shift as a result of the collision is very small. Thus the wave scattered from the ℓ-th particle, which can be expressed as a spherically outgoing wave in the asymptotic region $(R_\ell/r \ll 1)$, is

$$\psi_\ell = -\frac{a_\ell A}{r_\ell} e^{i(k_f r_\ell + \underset{\sim}{k}_i \cdot \underset{\sim}{R}_\ell - \omega_i t)}$$

$$\cong -\frac{a_\ell A}{r} e^{i(k_f r + \underset{\sim}{\kappa} \cdot \underset{\sim}{R}_\ell - \omega_i t)}, \qquad \text{(II.2)}$$

where $\underset{\sim}{\kappa} = \underset{\sim}{k}_i - k_f \hat{\underset{\sim}{r}}$ and r_ℓ, to a good approximation, is $r - \hat{\underset{\sim}{r}} \cdot \underset{\sim}{R}_\ell$. The strength of the interaction is given by $-a_\ell$, the scattering amplitude at $r = 1$ cm if amplitude of the incident wave is unity.+ We note that in the usual quantum interpretation the magnitude of scattered neutron momentum k_f is related to the energy ω_f, which is not necessarily equal to ω_i since $\underset{\sim}{R}_\ell$ may contain additional time dependence. Only if the particle is stationary is $\omega_f = \omega_i$ and $k_f = k_i$, and the scattering is said to be elastic.

+ It should be noted that we have assigned a scattering length to a given nucleus, and in so doing have avoided all questions concerning details of the nuclear forces. The situation is very much like the scattering of water waves by vertical posts made of different materials. The scattering amplitudes of the various posts will be different, and in principle can be calculated if the necessary theory were available. In the absence of such a theory the alternative solution will be to measure the different scattering amplitudes. Our attitude follows essentially the latter point of view; that is to say, we shall characterize the ability of a nucleus in scattering neutrons by the scattering length a, an experimentally determined quantity. The negative sign is chosen to agree with convention. Interested reader may see J. M. Blatt and V. Weisskopf, Theoretical Nuclear Physics, Wiley (1952).

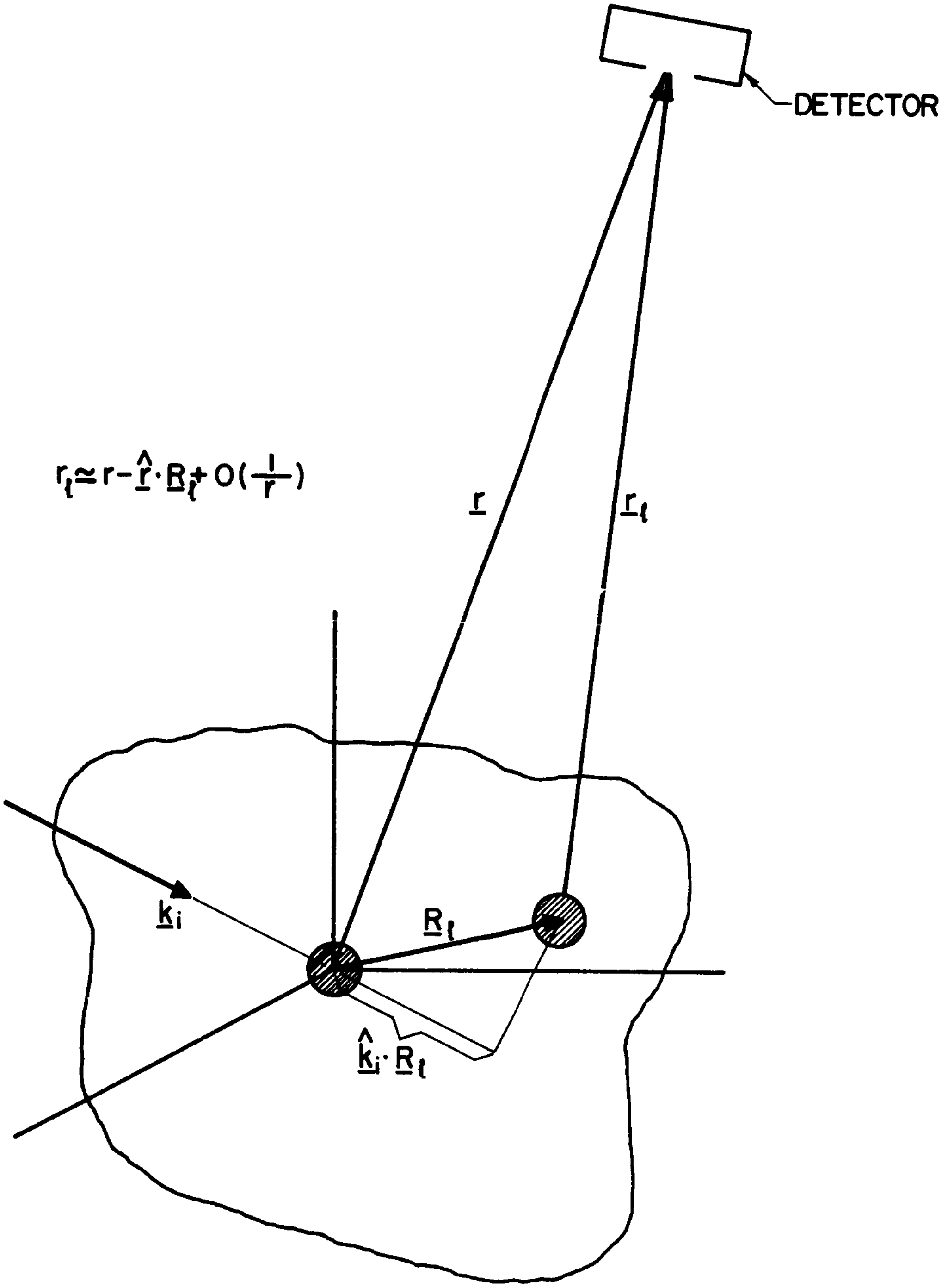

Figure 2. Coordinates for Neutron Scattering by Particles.

The total scattered wave is a superposition of waves scattered by all particles,

$$\psi = -\frac{A}{r}\, e^{-i\omega_i t} \sum_{\ell}^{N} a_\ell e^{i(k_f r_\ell + \underset{\sim}{\kappa}\cdot \underset{\sim}{R}_\ell)} , \qquad \text{(II.3)}$$

where k_f is understood to depend upon the time dependence of $\underset{\sim}{R}_\ell$. For the moment we consider all the particles to be held fixed. In the absence of particle motion the factor $e^{ik_f r} = e^{ik_i r}$ can be taken outside the summation, and the differential cross section is found to be

$$\sigma d\Omega = \frac{1}{N} \left| \sum_{\ell}^{N} a_\ell e^{i\underset{\sim}{\kappa}\cdot\underset{\sim}{R}_\ell} \right|^2 .$$

It will be shown that for crystals the summation of the various phase factors leads to the well-known Bragg condition for interference scattering, a result of superposing waves scattered from ordered arrays of crystal planes. Of course, in the evaluation, terms for which $\ell = \ell'$ constitute the contribution to direct scattering, a result of single particle effects. In the special case of only one scatterer present, the total cross section becomes

$$\sigma = \int \sigma d\Omega = 4\pi a^2 ,$$

where a is often referred to as the bound-atom scattering length, and scattering is seen to be isotropic.

In order to study the dependence of cross section upon dynamics of the scattering system we consider a hypothetical simple cubic, monatomic crystal containing N nuclei with zero spin, each executing independent oscillations about its equilibrium position (lattice site). The instantaneous position of the ℓ-th particle becomes $\underset{\sim}{R}_\ell(t) = \underset{\sim}{x}_\ell + \underset{\sim}{u}_\ell(t)$, where $\underset{\sim}{x}_\ell$ is the equilibrium position and $\underset{\sim}{u}_\ell(t)$ the instantaneous displacement from $\underset{\sim}{x}_\ell$,

$$\underset{\sim}{u}_\ell(t) = \underset{\sim}{A} \cos(\omega t - \Delta_\ell) \ . \qquad \text{(II.4)}$$

In this simple description all particles have the same amplitude A and fundamental vibrational frequency ω. The fact that the oscillations of any two particles are not coupled is expressed by the presence of an arbitrary phase Δ_ℓ. Using this time dependence we find that the scattered wave may be written as

$$\psi = -\frac{Ae^{-i\omega_i t}}{r} \sum_\ell^N a_\ell e^{i(k_f r + \underset{\sim}{\kappa}\cdot\underset{\sim}{x}_\ell)}$$

$$(x) \sum_{n=0}^{\infty} i^n J_n(\underset{\sim}{\kappa}\cdot\underset{\sim}{A})\{e^{in(\omega t-\Delta_\ell)} + e^{-in(\omega t-\Delta_\ell)}\} \ ,$$

since

$$e^{ix\cos y} = J_o(x) + 2\sum_{n=1}^{\infty} i^n J_n(x)\cos(ny) \ ,$$

which is readily obtained from the generating function of Bessel function of first kind. The leading term in the sum contains no time dependence and is seen to be the elastic part of the scattered wave while the remaining terms correspond to scattering with energy transfer (inelastic), the energy exchange for the n-th term being $\omega_f - \omega_i = \pm n\omega$. In computing the differential cross section we can ignore the cross terms. This is because in any physical measurement the intensity is actually integrated over an interval of time which is very large compared to time of flight or interaction time, hence all cross terms vanish. The cross section therefore becomes a sum of partial cross sections, each corresponds to a specific amount of energy transfer,

$$\sigma d\Omega = \sum_{n=0}^{\infty} \sigma^{\pm n} \, d\Omega \ ,$$

where upper and lower signs denote the increase or decrease of neutron energy by an amount $n\hbar\omega$. The discrete and uniform nature of the inelastic scattering is expected since in quantum theory an oscillator may undergo changes only in multiples of its level spacing $\hbar\omega$. The partial cross sections cannot be measured unless the measurement is performed with an energy-sensitive counter so that scattered neutrons with different energies may be differentiated. In order to exhibit the energy exchange in a scattering process more explicitly let us define a differental cross section $\sigma d\Omega dE_f$, which depends upon final neutron energy, as

$$\sigma d\Omega = \int_{E_f} \sigma d\Omega dE_f \ ,$$

with

$$\sigma d\Omega dE_f = \sum_{n=0}^{\infty} \sigma^{\pm n} \, \delta(E_f - E_i \mp n\hbar\omega) d\Omega dE_f \quad ,$$

where $\delta(x)$ is the Dirac delta. It is to be understood that the interval of energy integration depends upon the detection system. If the finite resolution of the instrument is less than the level spacing then $\sigma^{\pm n} d\Omega$ can be obtained for any given n. On the other hand, if the counter is not energy-sensitive then only $\sigma d\Omega$, which contains elastic as well as all the inelastic contributions, will be measured. For our purposes, it will be sufficient to confine our attention to two lowest-order processes,

$$\sigma^{o} d\Omega dE_f = \frac{1}{N} \, \delta(E_f - E_i) J_0^2(\underset{\sim}{\kappa} \cdot \underset{\sim}{A}) \left| \sum_{\ell} a_\ell e^{i\underset{\sim}{\kappa} \cdot \underset{\sim}{x}_\ell} \right|^2 d\Omega dE_f \ ,$$

$$\sigma^{\pm 1} d\Omega dE_f = \frac{1}{N} \, \delta(E_f - E_i \mp \hbar\omega) J_1^2(\underset{\sim}{\kappa} \cdot \underset{\sim}{A}) \sum_{\ell} a_\ell^2 d\Omega dE_f \quad .$$

We note that interference effects do not contribute to first-order inelastic scattering, a result equally valid for $n > 1$. This is a consequence of averaging over all arbitrary phases Δ_ℓ and the lack of interference is expected since there is no correlation among the various vibrations. The effects of such correlation are seen in the following discussion.

We next introduce additional complexity into our analysis by treating the particle oscillations in terms of traveling waves. It will be shown later that under certain reasonable assumptions regarding the inter-particle potential one can write in general

$$\underset{\sim}{u}_\ell(t) = \sum_\lambda \underset{\sim}{A}_\lambda \cos\{\underset{\sim}{k}_\lambda \cdot \underset{\sim}{x}_\ell - \omega_\lambda t\} , \tag{II.5}$$

where $\underset{\sim}{k}$ is the propagation vector and summation over λ is actually a double sum, one involving the N allowed values of $\underset{\sim}{k}$ and another the three values of ω for each given $\underset{\sim}{k}$. As before we obtained the elastic differential cross section as

$$\sigma^0 d\Omega dE_f = \frac{1}{N} \delta(E_f - E_i) \left|\sum_\ell a_\ell e^{i\underset{\sim}{\kappa}\cdot\underset{\sim}{x}_\ell}\right|^2 \left[\prod_\lambda J_0(\underset{\sim}{\kappa}\cdot\underset{\sim}{A}_\lambda)\right]^2 d\Omega dE_f .$$

We note that $J_0(\underset{\sim}{\kappa}\cdot\underset{\sim}{A}_\lambda)$ should be averaged over a thermal distribution of λ-th oscillation mode.(3) This is because if we regard A_λ as the amplitude of the λ-th oscillator whose energy is $\frac{1}{2} A_\lambda^2 \omega_\lambda^2$ then there exists a distribution of A_λ given by

$$\begin{aligned} P(A_\lambda)dA_\lambda &= P(E_\lambda)dE_\lambda \\ &= e^{-A_\lambda^2\omega_\lambda^2/2k_oT} \left(\int_0^\infty e^{-A_\lambda^2\omega_\lambda^2/2k_oT} A_\lambda dA_\lambda\right)^{-1} A_\lambda dA_\lambda , \end{aligned}$$

where k_o is Boltzmann constant and T the temperature of the system. Hence

$$\begin{aligned} \langle J_0(\underset{\sim}{\kappa}\cdot\underset{\sim}{A}_\lambda)\rangle &= \int_0^\infty J_0(\underset{\sim}{\kappa}\cdot\underset{\sim}{A}_\lambda) P(A_\lambda) dA_\lambda \\ &= e^{-k_oT(\underset{\sim}{\kappa}\cdot\hat{\underset{\sim}{A}}_\lambda)^2/2\omega_\lambda^2} , \end{aligned}$$

which can be readily obtained by using the series representation of J_o and integrating term by term. It is interesting to observe that this factor strongly attenuates the scattering at high temperature and large momentum transfer and is unity at $T = 0$. Since the average energy of the oscillator is k_oT by equipartition then we may replace the exponent by $-(\underset{\sim}{\kappa}\cdot\underset{\sim}{\hat{A}}_\lambda)^2 \langle A_\lambda^2\rangle/4$, where $\langle A_\lambda^2\rangle$ is average of the amplitude squared. The elastic cross section now becomes

$$\sigma^o d\Omega dE_f = \frac{1}{N}\,\delta(E_f - E_i)e^{-2W}\Big|\sum_\ell a_\ell e^{i\underset{\sim}{\kappa}\cdot\underset{\sim}{x}_\ell}\Big|d\Omega dE_f \ ,$$

where $W = \frac{1}{2}\sum_\lambda(\underset{\sim}{\kappa}\cdot\underset{\sim}{\hat{A}}_\lambda)^2\langle A_\lambda^2\rangle$. The factor e^{-2W} is known as the Debye-Waller factor, originally derived in X-ray diffraction to account for temperature effects. We will obtain the quantum analogues of this factor in a later calculation.

Unfortunately the inelastic cross sections are not as readily obtained. Since the complete problem will be discussed in the quantum treatment we can best avoid excessive manipulation by considering a special case. Suppose we are only interested in the scattering produced by a particular mode of oscillation, say the λ-th mode, then we may effectively regard all the other terms in (5) to be small by comparison and immediately obtain

$$\sigma_\lambda^{\pm1} d\Omega dE_f = \frac{1}{4N}\,\delta(E_f - E_i \mp \hbar\omega_\lambda)e^{-2W}(\underset{\sim}{\kappa}\cdot\underset{\sim}{A})^2\langle A_\lambda^2\rangle$$

$$(x)\ \Big|\sum_\ell a_\ell e^{i(\underset{\sim}{\kappa}\mp\underset{\sim}{k}_\lambda)\cdot\underset{\sim}{x}_\ell}\Big|^2\, d\Omega dE_f \ ,$$

where we have used the small argument representation,

$$J_n(x) \simeq \frac{1}{n!}\left(\frac{x}{2}\right)^n \ .$$

It is noted that inelastic interference effects are present due to (5) in which all oscillations are dynamically coupled. Also one sees that the cross section varies as the square of momentum transfer and inversely as the square of vibrational frequency. Finally we note that the above results are independent of whether energy is gained or lost, which, as we will see, is a direct consequence of classical analysis.

Thus far we have attempted to show that certain qualitative aspects of neutron scattering can be illuminated by a simplified semi-classical approach. The treatment is admittedly not rigorous nor are the cases examined the most realistic and sophisticated models one can construct, nevertheless, they do lead to results pertinent to the understanding of the physics involved and provide a systematic comparison with the rigorous quantum mechanical calculation given in the later sections.

III. GENERAL FORMULATION OF NEUTRON SCATTERING BY MACROSCOPIC SYSTEMS

We will now derive the quantum formalism which will be used in subsequent calculations to predict results that can be correlated with information obtained from neutron-diffraction experiments. In the description of the collision process it is not necessary to specify the physical state of the scatterer so that the formulation is equally valid whether the scattering system is in gaseous, liquid, or solid state; later applications will, of course, be restricted to neutron interactions with crystal lattices. It will be seen that the present analysis does not include neutron-nuclear spin interaction. The effects due to presence of nuclear spins will be discussed separately in a later section.

Consider again the idealized experiment of Section II where

$$\sigma d\Omega = \frac{\underset{\sim}{J}_f \cdot \hat{\underset{\sim}{r}}}{N \underset{\sim}{J}_i \cdot \hat{\underset{\sim}{Z}}} \; r^2 d\Omega \; .$$

In quantum theory it is well known that the particle current is given by

$$\underset{\sim}{J} = \frac{i\hbar}{2m} \{(\underset{\sim}{\nabla}\psi^+)\psi - \psi^+(\underset{\sim}{\nabla}\psi)\} \; ,$$

where ψ is the particle wave function. In order to obtain the neutron wave function we characterize the scattering process by the stationary Schrödinger equation

$$H\Psi = \mathcal{E}\Psi, \tag{III.1}$$

where $\mathcal{E}$ is the total energy of the system, scatterer plus neutron, whose wave function is denoted as Ψ. The appropriate Hamiltonian is

$$H = \underset{\sim}{p}^2/2m + H_s + V \; ,$$

where $\underset{\sim}{p}^2/2m$ is the kinetic energy of the neutron, H_s the Hamiltonian of the scattering system, and V the neutron-nuclear interaction potential. It is expected that given a system whose Hamiltonian is H_s, there exists a complete, orthonormal set of functions $\{\Phi_n\}$ which satisfy $H_s\Phi_n = \epsilon_n\Phi_n$, where ϵ_n is the eigenvalue corresponding to the n-th eigenstate. Then we can write

$$\Psi(\underset{\sim}{r},\underset{\sim}{R}) = \sum_n \psi_n(\underset{\sim}{r})\Phi_n(\underset{\sim}{R}) ,$$

where $\underset{\sim}{r}$ represents neutron position and $\underset{\sim}{R} = \{\underset{\sim}{R}_1,\underset{\sim}{R}_2,\ldots,\underset{\sim}{R}_n\}$ represents the set of $3N$ coordinates of a system containing N particles. $\psi_n(\underset{\sim}{r})$ is seen to be that neutron state which corresponds to the n-th state of the scattering system.

Inserting the above expansion into (1), we obtain

$$\frac{\underset{\sim}{p}^2}{2m}\psi_n + \sum_{n'} \langle n|V|n'\rangle\psi_{n'} = (\mathcal{E} - \epsilon_n)\psi_n ,$$

where

$$\langle n|V|n'\rangle = \int d\underset{\sim}{R}\,\Phi_n^*(\underset{\sim}{R})V\Phi_n(\underset{\sim}{R}) .$$

Writing the linear momentum operators $\underset{\sim}{p}$ as $\frac{\hbar}{i}\underset{\sim}{\nabla}$,

$$(\underset{\sim}{\nabla}^2 + k_n^2)\psi_n = \frac{2m}{\hbar^2}\sum_{n'}\langle n|V|n'\rangle\ \psi_{n'} , \qquad \text{(III.2)}$$

where

$$k_n^2 = \frac{2m}{\hbar^2}E_n , \quad E_n = \mathcal{E} - \epsilon_n .$$

Equation (2) represents a system of integro-differential equations and is best treated in the form of integral equations through the use of

Green's function. If we observe that

$$(\nabla^2 + k_n^2)G_n(\mathbf{r},\mathbf{r}') = -\delta(\mathbf{r},\mathbf{r}')$$

$$(\nabla^2 + k_n^2)\psi_n^o(\mathbf{r}) = 0 \; , \qquad \text{(III.3)}$$

then because (2) is a linear equation,

$$\psi_n(\mathbf{r}) = \psi_n^o(\mathbf{r}) - \frac{2m}{\hbar^2}\sum_{n'} \int d\mathbf{r}' G_n(\mathbf{r},\mathbf{r}')\langle n|V|n'\rangle \psi_{n'}(\mathbf{r}') .$$

Physically, if there were no interaction between the neutron and scatterer then we would expect the wave function to be just the product of the incident neutron and initial system wave functions. Hence,

$$\psi_n^o(r) = \delta_{nn_o} e^{ik_i \cdot r}$$

where n_o is the initial state of the scattering system. The first equation in (3) can be solved[10] to give

$$G_n(r,r') = \frac{e^{ik_n|\mathbf{r}-\mathbf{r}'|}}{4\pi|\mathbf{r}-\mathbf{r}'|} \; .$$

Since in the region where the neutrons are counted $\frac{r}{r'} \gg 1$, terms of order r^{-2} and higher can be ignored. Then

$$G_n(\mathbf{r},\mathbf{r}') \cong \frac{e^{ik_n r}\, e^{-i\mathbf{k}_n \cdot \mathbf{r}'}}{4\pi r} \; ,$$

and

$$\psi_n(\mathbf{r}) \cong \delta_{nn} e^{ik_i \cdot r} - \frac{2m}{4\pi\hbar^2}\frac{e^{ik_n r}}{r}\sum_{n'} \int d\mathbf{r}' e^{-ik_n \cdot \mathbf{r}'}\langle n|V|n'\rangle \psi_{n'}(\mathbf{r}') . \qquad \text{(III.4)}$$

According to the "First Born Approximation", the solution to (4) is obtained by setting $\psi_{n'}(\mathbf{r}') = \delta_{n'n_o} e^{ik_i \cdot \mathbf{r}'}$; in other words we iterate once to get

$$\psi_n(\underset{\sim}{r}) \cong \delta_{nn_o} e^{i\underset{\sim}{k}_i \cdot \underset{\sim}{r}} + f_{nn_o}(\underset{\sim}{\Omega}) \frac{e^{ik_n r}}{r} \quad . \tag{III.5}$$

The first term in (5) is the unscattered incident beam and the second term can be interpreted as the scattered spherically outgoing wave whose amplitude is

$$f_{nn_o}(\underset{\sim}{\Omega}) = - \frac{m}{2\pi\hbar^2} \int_{r'} dr' e^{i(\underset{\sim}{k}_i - \underset{\sim}{k}_n)\cdot \underset{\sim}{r}'} \langle n|V|n_o\rangle . \tag{III.6}$$

Now the differential scattering cross section for the corresponding neutron initial and final momenta $\underset{\sim}{k}_i$ and $\underset{\sim}{k}_n$, and system initial and final states n_o and n becomes

$$\sigma_{n_o n} d\Omega = \frac{k_n}{Nk_o} |f_{nn_o}|^2 d\Omega \ . \tag{III.7}$$

The cross section is written for specific initial and final states. Usually the final state of the scattering system is not observed so we will sum over all possible final states,

$$\sigma d\Omega = \frac{d\Omega}{N} \sum_{n_o n} P_{n_o} \frac{k_n}{k_i} |f_{nn_o}|^2 \ , \tag{III.8}$$

where P_{n_o} is the probability that the scatterer is initially in state n_o. Should the initial state be prepared P_{n_o} becomes a kronecker delta.

It is obvious that the statement of energy conservation for the problem under consideration is

$$E_n + \epsilon_n = E_i + \epsilon_{n_o} \ ,$$

where $E = \frac{(\hbar k)^2}{2m_o}$ and ϵ are the energies of neutron and scatterer respectively. This condition can be explicitly incorporated into the cross section by defining as before

$$\int_{E_f} \sigma d\Omega dE_f = \sigma d\Omega ,$$

and noting that

$$\delta(x) = \frac{1}{2\pi\hbar} \int_{-\infty}^{\infty} dt e^{itx/\hbar} ,$$

the latter being merely an integral representation of the Dirac delta-function. Now we obtain as an expression for the energy- and angle-dependent differential scattering cross section

$$\sigma d\Omega dE_f = d\Omega dE_f \frac{k_f}{2\pi\hbar N k_i} \int dt e^{\frac{-it}{\hbar}(E_f - E_i)}$$

$$(x) \sum_{nn_o} P_{n_o} e^{\frac{-it}{\hbar}(\epsilon_n - \epsilon_{n_o})} |f_{nn_o}|^2 ,$$

where on account of the delta function k_n, E_n can be written as k_f, E_f; this is to avoid confusion and to indicate that the scattered neutron energy is an observed quantity. The above manipulation conveniently removes the factor k_n/k_i from the summand.

It is noted that the amplitude of the scattered wave contains the neutron-nuclear interaction potential V. For sufficiently slow neutrons such that the wavelength is large compared to the range of nuclear forces ($E \lesssim$ ev) a neutron-nuclear collision can be characterized quite accurately as a "localized impact" for which the interaction potential is the "Fermi pseudo-potential",

$$V_{sn} = \frac{2\pi\hbar^2}{m_o} \sum_{\ell}^{N} a_\ell \delta(\underset{\sim}{r} - \underset{\sim}{R}_\ell) , \qquad \text{(III.10)}$$

where a_ℓ is the scattering length of the ℓ-th particle. The particular form of the potential is so chosen that the use of "First Born Approximation" will give the correct total scattering cross section for a free atom, $\sigma = 4\pi a_f^2$, where a_f is the experimentally determined free-atom scattering length. The relation between a_f and the scattering length a used in (10) is seen from the following argument. In an analysis of two-particle collision it is natural to use the relative coordinates in which the mass factor appears as a reduced mass $\mu = \frac{m_o M}{m_o+M}$. Then we choose the potential as

$$V_{sn} = \frac{2\pi\hbar^2}{\mu} a_f \delta(\underset{\sim}{r}-\underset{\sim}{R})$$

in order to make the scattering amplitude independent of mass. Now if the system contains more than one scatterer there is no advantage in defining a center of mass and in this case we can write

$$V = \frac{2\pi\hbar^2}{m_o} a_f \delta(\underset{\sim}{r}-\underset{\sim}{R})$$

with the bound-atom scattering length defined as $a = \frac{(m_o+M)}{M} a_f$. With this choice of V the scattered amplitude takes the form

$$f_{nn_o} = - \sum_{\ell} \langle n | a_\ell e^{-i\underset{\sim}{\kappa}\cdot\underset{\sim}{R}_\ell} | n_o \rangle \quad ,$$

where $\underset{\sim}{\kappa} = \underset{\sim}{k}_i - \underset{\sim}{k}_f$. The summation over n in (9) becomes

$$\sum_n e^{\frac{-it}{\hbar}(\epsilon_n-\epsilon_{n_o})} |f_{nn_o}|^2 = \sum_{\ell\ell'} a_{\ell'} a_\ell \langle n_o | \Lambda_{\ell'}^{\dagger}(t) \Lambda_\ell | n_o \rangle ,$$

where

$$\Lambda_\ell = e^{-i\underset{\sim}{\kappa}\cdot\underset{\sim}{R}_\ell}$$

$$\Lambda_\ell(t) = e^{\frac{iH_s t}{\hbar}} \Lambda_\ell e^{\frac{-iH_s t}{\hbar}} = e^{-i\underset{\sim}{\kappa}\cdot(e^{\frac{iH_s t}{\hbar}} \underset{\sim}{R}_\ell e^{\frac{-iH_s t}{\hbar}})} = e^{-i\underset{\sim}{\kappa}\cdot\underset{\sim}{R}_\ell(t)} ,$$

the time-independent and time-dependent dynamical variables being designated

R_ℓ and $R_\ell(t)$ respectively.(4) The time-dependent operator $R_\ell(t)$ satisfies the Heisenberg equation of motion

$$i\hbar \frac{dR_\ell(t)}{dt} = [R_\ell(t), H_s] .$$

Equation (9) finally becomes

$$\sigma d\Omega d\rho = d\Omega d\rho \frac{k_f}{2\pi N k_i \hbar} \int dt e^{-i\rho t}$$

$$(x) \sum_{n_o \ell \ell'} P_{n_o} a_\ell a_{\ell'} \langle n_o | \Lambda^\dagger_{\ell'}(t) \Lambda_\ell | n_o \rangle , \qquad \text{(III.11)}$$

where $\rho = (E_f - E_i)/\hbar$. This then is a convenient expression of differential scattering cross section for subsequent calculations. It is quite general since the macroscopic system has not been specified and should be valid as long as the incident neutron is in the energy range for which nuclear scattering is isotropic in the center of mass coordinate system and independent of neutron energy.

IV. THE EINSTEIN CRYSTAL

In this section we wish to illustrate some general aspects of neutron scattering by crystals without applying the quantum formalism to a complicated model and thus getting involved in a great deal of calculations. It will be sufficient for our purpose to limit our considerations to a simple description of particle interactions -- the Einstein crystal. For although this model is a severe idealization of an actual crystal, nevertheless, as we will presently see, it is capable of giving most of the essential features of neutron scattering which can be obtained by an analysis based upon a considerably more realistic model. We will not give any details of the calculation leading to the scattering cross section since it is completely analogous to that for the more general crystal model to be discussed in Section VII. Instead we will examine the result and obtain some limiting cases which are essential to the understanding of the scattering process.

For the Einstein crystal it is assumed that each particle (nucleus) sees the same surroundings and therefore executes independent, isotropic oscillations about an equilibrium position, its lattice site. In this manner it is reasonable to assume that all the fundamental vibrational frequencies are identical. We note that in the case of polyatomic crystal where mass differences among the different particles are large it is possible that the vibrational motion of the light atoms can be adequately described by the above model. Such an example could be the hydrogen atoms in zirconium hydride.

Since the quantity of interest is the energy- and angle-dependent differential cross section we see from the previous section that the calculation involves first obtaining the matrix elements $\langle n|\Lambda^{+}_{\ell'}(t)\Lambda_{\ell}|n\rangle$ which in turn requires a determination of the eigenstates $|n\rangle$. Accordingly we consider a monatomic crystal containing N particles which are labelled by subscript ℓ. In view of the adopted dynamical model, the Hamiltonian for the system is that of 3N independent harmonic oscillators, i.e.,

$$H_s = \frac{1}{2} \sum_{\ell=1}^{N} \sum_{\alpha=1}^{3} \left[\frac{P^2_{\ell\alpha}}{M} + M\omega^2 u^2_{\ell\alpha}\right] , \tag{IV.1}$$

where $P_{\ell\alpha} = \frac{\hbar}{i} \frac{\partial}{\partial u_{\ell\alpha}}$ and subscript α denotes the α-th component of a vector. As before, $\underset{\sim}{u}_{\ell}$ is the instantaneous displacement of the ℓ-th particle from its equilibrium position $\underset{\sim}{x}_{\ell}$, i.e., $\underset{\sim}{R}_{\ell} = \underset{\sim}{x}_{\ell} + \underset{\sim}{u}_{\ell}$. Having specified the scattering system we can proceed to obtain the cross section using equation (III.11). The differential scattering cross section per unit solid angle and unit neutron final energy is

$$\begin{aligned} \sigma = {} & \delta(\rho)e^{-D\kappa^2} \{a_1^2 I_o(P\kappa^2) - a_2^2\} \\ & + \delta(\rho)a_2^2 e^{-D\kappa^2} \frac{1}{N}|\sum_{\ell} e^{i\kappa\cdot x_{\ell}}|^2 \\ & + \frac{k_f}{k_i} a_1^2 e^{-D\kappa^2} \{\sum_{n=1}^{\infty} I_n(P\kappa^2)[e^{n\nu}\, \delta(\rho+n\omega) + e^{-n\nu}\, \delta(\rho-n\omega)]\}, \end{aligned} \tag{IV.2}$$

where $D = \frac{\hbar}{2M\omega} \coth(\nu)$, $P = \frac{\hbar}{2M\omega} \operatorname{csch}(\nu)$, $\nu = \frac{\hbar\omega}{2k_o T}$,

and k_o is the Boltzmann constant. Other quantities appearing in this equation will be defined in the following discussion.

In Equation (2) we note that the delta functions represent conditions of energy conservation. The two terms proportional to $\delta(\rho)$ give the elastic contribution while terms containing $\delta(\rho \pm n\omega)$ constitute the inelastic portion of the cross section, upper and lower signs corresponding respectively to neutron loss and gain of energy by an amount $n\hbar\omega$. The uniform and discrete nature of the energy transfer is expected since an oscillator can undergo energy exchange only in multiples of its level spacing. We observe that the cross section contains all the inelastic processes for which energy conservation can be satisfied, and that at any finite temperature neutron energy loss is more probable than neutron energy gain.+ The two processes become comparable when $k_oT >> n\hbar\omega$ which is the semi-classical result of Section II. It is interesting to note that neutrons cannot gain any energy from the crystal at $T = 0$ because $e^{-n\nu}$, which can be interpreted as a measure of the probability of finding the oscillator in the n-th eigenstate, vanishes. For large vibrational frequency we may use the small argument representation of the modified Bessel function

$$I_n(x) \simeq \left(\frac{x}{2}\right)^n \frac{1}{n!} \quad ,$$

so that in the limit as ω becomes infinite all inelastic terms vanish. Physically this corresponds to the situation in which all particles are rigidly fixed at the lattice sites so the crystal cannot possibly interchange energy with the neutron. Elastic scattering, however, is still allowed.

The elastic contributions to the cross section are exhibited in two separate terms. The second term contains the interference factor

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+ Of course, the neutron may lose energy to the crystal lattice only if its energy is greater than the oscillator level spacing.

$$\frac{1}{N} \left|\sum_{\ell} e^{i\kappa\cdot x_\ell}\right|^2 \quad .$$

It is seen that lattice symmetries affect only the interference part of the scattering and that interference effects are purely elastic processes. That the latter is a direct consequence of a crystal model which assumes uncorrelated particle motion has already been mentioned in Section II. The more realistic model which we will describe in the next section is one in which all particle motions are correlated, and for that case it will be seen, as in Section II, that inelastic interference scattering is indeed permissible.

The attentuating exponential factor $e^{-D\kappa^2}$ is the quantum analogue of the Debye-Waller factor. At high temperature or small ν it is seen that all processes are appreciably attentuated; the effect does not vanish entirely at $T = 0$, apparently due to the oscillator zero point energy. On the other hand, for sufficiently small ν $P\kappa^2$ will be such that we may use the asymptotic form of the modified Bessel function, $I_n(x) \simeq (2\pi x)^{-1/2} e^x$, the exponential part of which cancels the Debye-Waller factor. Obviously the same situation holds for the case of large κ^2 instead of large T, for this reason we see that in the region of high momentum transfer interference effects will be negligible.

We next consider the scattering lengths a_1 and a_2 which appear in Equation (2). The necessity for the distinction arises from the fact that the scattering system may contain a mixture of isotopes for which the scattering lengths differ or the interaction potential may be spin-dependent.+ For the present it suffices to mention that a_1 and a_2 are the appropriately averaged scattering lengths for those terms with $\ell = \ell'$

+ Isotopic mixture is discussed in Section VII while spin dependence is treated in Section VIII.

and $\ell \neq \ell'$ respectively [see Equation (III.11)], and in this sense terms in (2) containing a_1^2 and a_2^2 may be identified as direct and interference scattering. With this interpretation we observe that the two processes are influenced by independent aspects of the scattering system, the former being only sensitive to lattice dynamics whereas the latter is governed by lattice symmetries. In the presence of isotopic mixture and spin-dependent interaction the scattering lengths are

$$a_1^2 = \left< \frac{I+1}{2I+1} a_+^2 \right> + \left< \frac{I}{2I+1} a_-^2 \right>$$

$$a_2^2 = \left< \frac{I+1}{2I+1} a_+ + \frac{I}{2I+1} a_- \right>^2$$

where I is the spin of the nucleus and $< >$ denotes isotopic average. Scattering lengths a_+ and a_-, defined in Section VII, are those for which neutron spin is respectively parallel and antiparallel to nuclear spin. When spin effects are absent, $I \to 0$ and we have $a_1^2 = \langle a^2 \rangle$ and $a_2^2 = \langle a \rangle^2$, where a is the ordinary scattering length introduced in Equation (III.10). Furthermore, if the scattering system is monoisotopic then $a_1^2 = a_2^2 = a^2$.

We are now in a position to examine the qualitative behavior of the scattering cross section. Since among a large number of the experimental results the measured quantity is independent of scattering angle and neutron final energy, it is only necessary to discuss the dependence upon neutron initial energy of the total cross section, which is obtained by integrating (2) over all final energies and directions of scattering. We note that the scattering samples used in these experiments are polycrystalline so that the interference term, which depends upon crystal orientation, should be averaged over all crystal orientations. However, this aspect is not important for the discussion in this section and will be treated in Section VII. Let us therefore consider the scattering cross section for a given incident neutron energy which can be measured, say, by a transmission experiment in which we will neglect spin and isotope effects.

For very low-energy neutrons ($E_i \lesssim .001$ ev) it is seen that the model predicts no elastic processes. This is because $I_0(x)$ is essentially unity which enables the cancellation of direct scattering, and the neutron wavelength is sufficiently long that the Bragg interference condition cannot be satisfied at any scattering angle. Also, in this region neutrons cannot lose energy; the only permissible inelastic process, therefore, is that in which neutrons gain energy and one can readily show that the cross section varies as $E_i^{-1/2}$ and increases with temperature. As the incident neutron energy is raised elastic processes begin to contribute; a significant increase occurs when the Bragg condition which allows the longest wavelength is just satisfied. At higher incident energies the interference term begins to be attentuated by the Debye-Waller factor. While the cross section will continue to exhibit sharp jumps as additional sets of crystal planes give rise to interference scattering the overall oscillatory behavior is damped. For the high-energy neutrons ($E_i \gtrsim$ ev) we can, therefore, expect no contribution from interference scattering; moreover, in this region one can neglect lattice binding and thermal effects and treat the scatterer as a free particle. The dominant inelastic process here is that in which neutrons loose energy since neutron energy gain becomes negligible because of the factor $e^{-\nu}$ with ν large. We obtain the limiting result for high energies , $\sigma = 4\pi a_f^2$. The above remarks are illustrated in Figure 3 which is in general agreement with observations for such scatterers as graphite, beryllium, and lead.[5] Finally we mention that beryllium, on account of its sharp Bragg cutoff and a low-energy cross section, which can be made even smaller upon cooling the material, has been used as a filter in experiments requiring very low-energy neutrons.[6]

We have just shown that the Einstein crystal can be used to understand certain general aspects of neutron scattering and to explain the behavior of the total scattering cross section. It is clear, however, that this simplified model cannot be adequate in predicting results which are sensitive to dynamical details of the scattering system. With the recent development of slow-neutron experiments as an important research tool in solid-state physics it is known that scattering measurements can be analyzed to yield valuable information on atomic motions and interparticle forces. Obviously, in order to do this there must exist a dynamical model for the crystal which treats particles dynamics to a sufficiently high degree of complexity and which still remains analytically tractable. The formulation of such a model will be considered next.

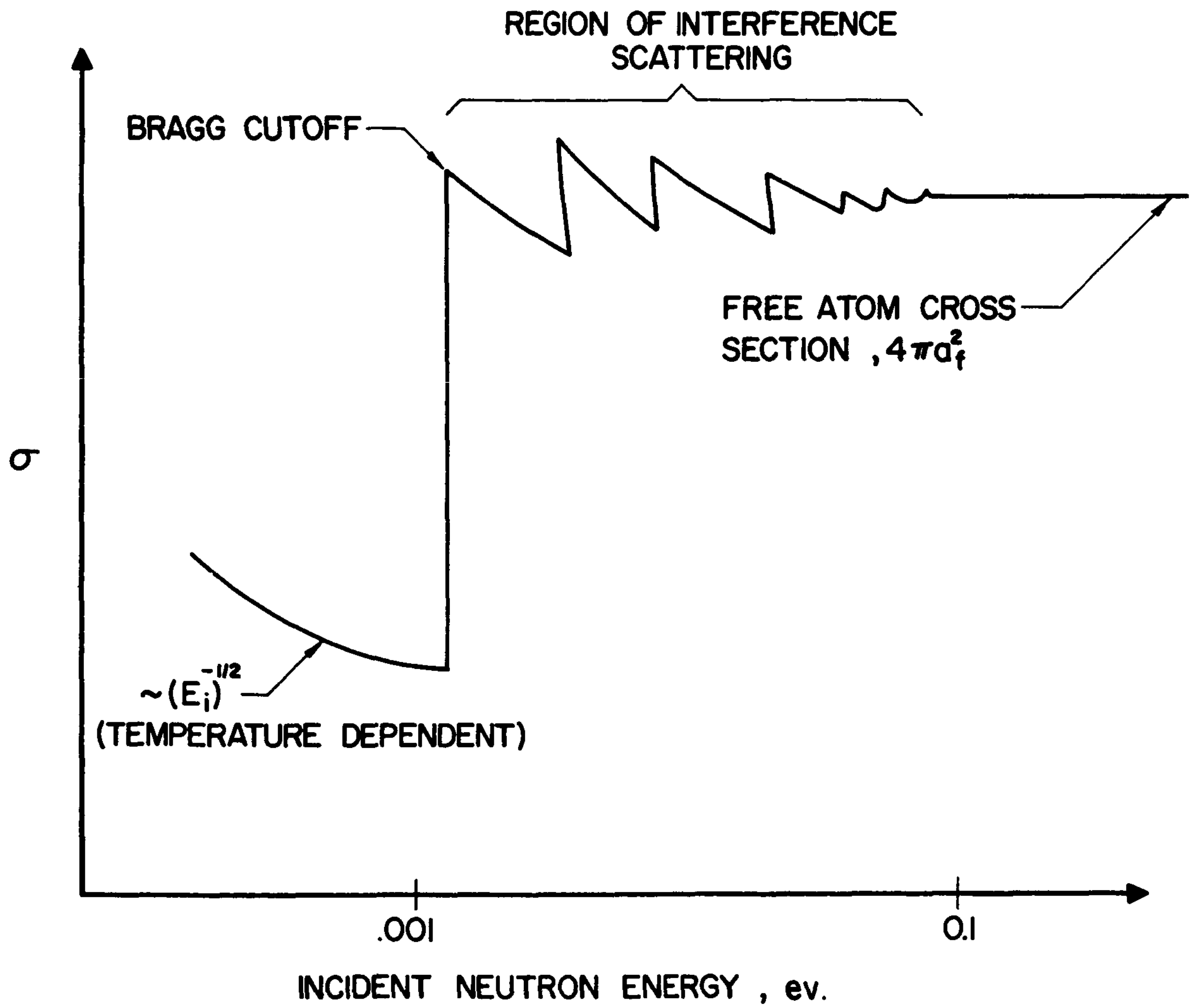

Figure 3. Qualitative Behavior of Total Scattering Cross Section.

V. A DYNAMICAL MODEL FOR CRYSTALS

As we have seen, the description of neutron interaction with atoms or molecules bound in a crystal depends to a large extent upon the energy of the neutron relative to that of the atoms in the lattice. For high-energy neutrons the lattice binding effect can be safely neglected in treating the collision process. The interval of time in which the neutron actually interacts with an atom will be small so that the atom can be effectively considered as being free, and if the wavelength of the neutron is sufficiently short the interference effects of the scattering from neighboring atoms will not be significant. It is then to be expected that any analysis in this region will not be sensitive to details of the scattering system and is of little interest in our discussion. On the other hand, when the neutron energy is in the region of .1 ev or less, the associated de Broglie wavelength is comparable to the interatomic distance for the lattice. Not only do the scattering atoms have to be treated collectively but their thermal motions and the effect of lattice binding will also have appreciable influence on the nature of the scattering. For this reason most scattering experiments are performed with very low-energy neutrons in order to study dynamical details and inter-atomic forces in crystals. In this section we will develop an appropriate model which will enable us to analyze these measurements. It will be more instructive to proceed classically, and later quantum analogues can be obtained for those results which are relevant to cross section calculations.

Consider a general crystal structure in which the ℓ-th lattice cell is located by a position vector $\underset{\sim}{x}_{\ell}$, whose components form a set of three integers ℓ^i appropriate to the lattice basic vectors $\underset{\sim}{a}_i$, $i = 1,2,3$. Let the n particles in each lattice cell be labeled s, $s = 1,2,\ldots,n$. As shown in Figure 4, the instantaneous position of the s-th particle in the ℓ-th cell is $\underset{\sim}{R}^s_\ell$, which can be written as

$$\underset{\sim}{R}^s_\ell = \underset{\sim}{x}^s_\ell + \underset{\sim}{u}^s_\ell$$

$$= \underset{\sim}{x}_\ell + \underset{\sim}{x}^s + \underset{\sim}{u}^s_\ell \ , \tag{V.1}$$

where u^s_ℓ is the instantaneous displacement from the equilibrium position x^s_ℓ. x^s is the equilibrium position vector of the s-th particle measured in the cell and is therefore independent of ℓ. For this particle the equations of motion are

$$M^s \frac{d^2}{dt^2} (u^s_{\ell\alpha}) = - \frac{\partial U}{\partial U^s_{\ell\alpha}} \ , \tag{V.2}$$

where we use Greek subscript α to denote a component of a vector and U represents the interparticle potential. Here M^s is the mass of the s-th particle. In the absence of detailed knowledge of U, the approximation conventionally employed is that proposed by Born and Oppenheimer.(7,8) For our purpose we shall adopt this approximation to obtain a realistic model for the crystal which still describes the particle motions as harmonic vibrations about their respective equilibrium positions. Compared to the Einstein crystal the nature of the motions is the same, but, as we will see, the additional complexity introduced lies in the fact that a large number of oscillation modes, which will no longer be uncorrelated, are allowed.

The Born-Oppenheimer method, when applied to the crystal, essentially consists of separating the Schrödinger equation for the system into

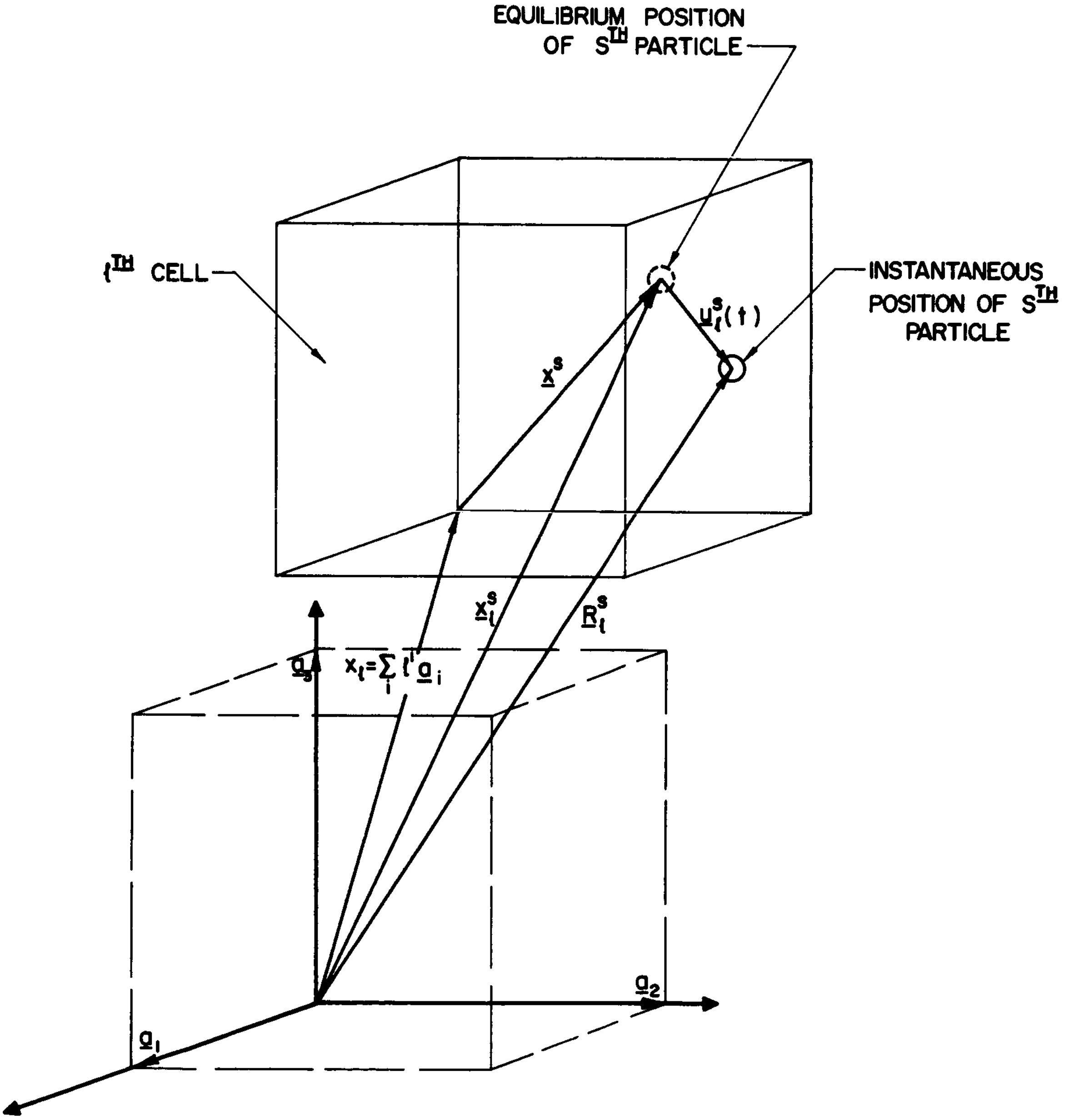

Figure 4. Position Vectors in a Simple Cubic Lattice.

two parts, one describing the electronic motion and the other the nuclear motion. It can be shown+ that the effective potential between nuclei is actually the eigenvalue of the electronic problem and in the harmonic approximation can be written as

$$U = \frac{1}{2} \sum_{\substack{\ell s \alpha \\ \ell' s' \alpha'}} U^{ss'}_{\ell\ell'\alpha\alpha'} \, u^{s}_{\ell\alpha} \, u^{s'}_{\ell'\alpha'} \;, \tag{V.3}$$

where the summation extends over all cells, particles and components, and

$$U^{ss'}_{\ell\ell'\alpha\alpha'} = \left[\frac{\partial^2 U}{\partial u^{s}_{\ell\alpha} \, \partial u^{s'}_{\ell'\alpha'}}\right]_{o} \;,$$

where subscript o means that the second-order derivative is to be evaluated at the equilibrium positions. Differentiating (3), we get++

$$\frac{\partial U}{\partial u^{s}_{\ell\alpha}} = \sum_{\ell' s' \alpha'} U^{ss'}_{\ell\ell'\alpha\alpha'} \, u^{s'}_{\ell'\alpha'} \;. \tag{V.4}$$

The equations of motion now become

$$M^{s} \frac{d^2 u^{s}_{\ell\alpha}}{dt^2} = - \sum_{\ell' s' \alpha'} U^{ss'}_{\ell\ell'\alpha\alpha'} \, u^{s'}_{\ell'\alpha'} \;, \tag{V.5}$$

where $U^{ss'}_{\ell\ell'\alpha\alpha'}$ can be interpreted as the α-th component of the force on particle (ℓ, s) due to a unit displacement of particle (ℓ', s') in the α'-direction. On account of lattice periodicity, this force is a function only of vector displacement and therefore can only depend upon the relative

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+ Readers not familiar with this particular aspect will find a thorough discussion in Reference 8, Chap. V.

++The same result is obtained if $\partial U/\partial u^{s}_{\ell\alpha}$ is simply expanded in a Taylor series in which only the first two terms are retained and noting that the first term represents the α-th component force on the particle (ℓ, s) in its equilibrium position and therefore vanishes.

cell index $\ell - \ell'$,

$$U^{ss'}_{\ell\ell'\alpha\alpha'} = U^{ss'}_{\ell-\ell'\alpha\alpha'} . \tag{V.6}$$

Equation (5) represents a system of simultaneous, linear differential equations and the set is infinite unless the cell indices are restricted. Since we are interested in actual macroscopic crystals the number of cells, no matter how large, will always be finite.+ If such a crystal were to contain more and more particles then the system approaches a continuum in the limit and the solution to the equation of motion should have the form of a solution to the wave equation, a traveling wave with infinite number of degrees of freedom. On the other hand, so long as the system remains discrete, the solution must contain the same number of degrees of freedom as the physical system. Accordingly, we seek a solution of the form

$$u^{s}_{\ell\alpha}(t) = (M^{s})^{-1/2}\, g^{s}_{\alpha} e^{i(2\pi k \cdot x^{s}_{\ell} - \omega t)} . \tag{V.7}$$

The usual oscillatory time dependence is chosen; the dependence of the circular frequency ω and its allowed values have yet to be determined. The appearance of the factor $\exp(2\pi i k \cdot x^{s}_{\ell})$ is perhaps not obvious. While this is closely analogous with the wave solution for an elastic continum there exists a somewhat subtle connection with the periodic property of the crystal lattice and a specific boundary condition. For fixed s and α the displacement is a periodic function of the cell location x_{ℓ}. Moreover, if the propagation vector k, unspecified as yet, is appropriately restricted, it is seen that the motions of particles occupying equivalent positions in cells

+ The use of (5) to describe a finite crystal implicitly assumes that surface effects are negligible. This is because (3) was derived for an infinite medium.

certain distance apart are described by the same solution. Therefore the range of $\underset{\sim}{k}$ can be so defined that the solution satisfies the boundary condition requiring particles on opposing surfaces of the crystal to move in unison. This boundary condition, which is actually expressed through the condition on $\underset{\sim}{k}$, does not prevent the description of the particle motions within the crystal by a superposition of a finite number of traveling waves as given by (7).+ At the same time, the solution is seen to exhibit the proper number of vibration modes. These remarks will be made more explicit after we discuss the determinations of frequency ω and coefficients g_{α}^{s}.

Inserting solution (7) into the equations of motion we obtain

$$\omega^2 g_{\alpha}^{s} = \sum_{s'\alpha'} C_{\underset{\sim}{k}\alpha\alpha'}^{ss'} \, g_{\alpha'}^{s'} \quad , \tag{V.8}$$

where

$$C_{\underset{\sim}{k}\alpha\alpha'}^{ss'} = \frac{e^{-2\pi i \underset{\sim}{k}\cdot(\underset{\sim}{x}^{s}-\underset{\sim}{x}^{s'})}}{\sqrt{M^s M^{s'}}} \sum_{\ell} U_{\ell\alpha\alpha'}^{ss'} \, e^{-2\pi i \underset{\sim}{k}\cdot \underset{\sim}{x}_{\ell}} \quad . \tag{V.8a}$$

Observe that without property (6), $C_{k\alpha\alpha'}^{ss'}$ would not be independent of the cell index. Equation (8) now represents $3n$ linear homogeneous equations in the $3\,n$ unknowns $g_{(\alpha)}^{s}$ since $s = 1,2,\ldots,n$ and $\alpha = 1,2,3$. For a given $\underset{\sim}{k}$, $C_{\underset{\sim}{k}\alpha\alpha'}^{ss'}$ is a $3n \times 3\,n$ matrix if (s,α) and (s',α') are considered as single indices. Of course, (8) will have non-trivial solutions only if the secular determinant vanishes, i.e.,

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+ The restricted motion of the surface particles is of no consequence since we have consistently neglected surface effects. One should be aware that these implications result directly from the form of solution chosen and the related boundary condition which, together with the interparticle potential, represent the crystal model we have adopted. Solution (7) with boundary condition, at best, describes only approximately the dynamical behavior of particles in an actual crystal.

$$|C^{ss'}_{\underset{\sim}{k}\alpha\alpha'} - \omega^2 \delta_{\alpha\alpha'} \delta_{ss'}| = 0 . \tag{V.9}$$

This is a $3n$ - degree equation in ω^2, and if the solutions are restricted to have only positive frequencies then both $\underset{\sim}{k}$ and $-\underset{\sim}{k}$ are admissible. Once the frequencies are known, they may be used in Equation (8) to obtain the coefficients g^s_α . According to (7) for a given $\underset{\sim}{k}$ there exists a real progressive wave corresponding to each of the $3n$ frequencies. However, not just any value of $\underset{\sim}{k}$ will lead to unique solutions. That this is a direct consequence of the assumed lattice periodicity property is seen in the following argument.

The solution (7) depends upon cell index ℓ only through the phase factor $\exp(2\pi i\underset{\sim}{k}\cdot\underset{\sim}{x}_\ell)$. We can define a set of reciprocal basic vectors $\underset{\sim}{b}^i$ related to the lattice basic vectors $\underset{\sim}{a}_i$ by

$$\underset{\sim}{b}^i = \frac{\underset{\sim}{a}_j \times \underset{\sim}{a}_k}{v_a} , \quad i,j,k \text{ in cyclic order,} \tag{V.10}$$

such that

$$\underset{\sim}{b}^i \cdot \underset{\sim}{a}_j = \delta_{ij} , \tag{V.11}$$

with $v_a = |\underset{\sim}{a}_1 \cdot \underset{\sim}{a}_2 \times \underset{\sim}{a}_3|$. The superscript serves to indicate that $\underset{\sim}{b}^i$ may be regarded as a set of contravariant vectors while $\underset{\sim}{a}_i$ will be the corresponding covariant set. Using these reciprocal basic vectors we define a reciprocal lattice in which a lattice vector is

$$\underset{\sim}{Y} = \sum_i y_i \underset{\sim}{b}^i , \tag{V.12}$$

where y_i are integers. According to (12), the scalar product between a reciprocal lattice vector $\underset{\sim}{Y}$ and a lattice vector $\underset{\sim}{x}_\ell$ is always an integer equal to $\sum_i y_i \ell^i$. Consequently the phase factor is unchanged if the propagation vector $\underset{\sim}{k}$, which when expressed in reciprocal lattice is not necessarily a reciprocal lattice vector, were changed by an amount equal to any reciprocal lattice vector $\underset{\sim}{Y}$. Furthermore, given a reciprocal lattice cell, a reciprocal lattice vector can always be chosen to connect any point outside the cell to a point inside. This implies that corresponding to every value of $\underset{\sim}{k}$ within the reciprocal lattice cell there exists a unique phase (and therefore unique solution) and corresponding to any value of $\underset{\sim}{k}$ outside the cell there exists no additional phase which is distinct. Hence, all the unique solutions are retained if $\underset{\sim}{k}$ is restricted to a reciprocal lattice cell.

The foregoing consideration of uniqueness of the solutions leads to a restriction of $\underset{\sim}{k}$. However, this does not completely specify $\underset{\sim}{k}$ since there exist a countably infinite number of $\underset{\sim}{k}$ values within the reciprocal lattice cell. This situation is to be expected since the physical boundary of the crystal has not yet been specified. Let us consider a crystal which contains N_i number of cells along the lattice basic vector $\underset{\sim}{a}_i$. For this case the solution to the equation of motion is that given by (7) where $\ell^i = 1,2,\ldots,N_i-1$. The boundary condition that particles on opposing surfaces of the crystal move in identical manner requires+ $u^s_{\ell\alpha} = u^s_{\ell_0\alpha}$, with $\underset{\sim}{x}_\ell - \underset{\sim}{x}_{\ell_0} = \sum_i \epsilon_i N_i a_i$, $\epsilon_i = 0,1$. This implies that

$$e^{2\pi i \underset{\sim}{k}\cdot(\underset{\sim}{x}^s_\ell - \underset{\sim}{x}^s_{\ell_0})} = e^{2\pi i \sum_{i=1}^{3} \epsilon_i k_i N_i a_i} = 1 \; , \qquad \text{(V.13)}$$

or $k_i N_i a_i$ = integer, where k_i are components of k along the unit

+ This boundary condition is known as the Born-Von Karman boundary condition originally proposed by Born and independently by Von Karman.

vectors $\underset{\sim}{a}_i/a_i$. It will be more convenient to express $\underset{\sim}{k}$ in terms of reciprocal basic vectors, i.e., $\underset{\sim}{k} = \underset{\sim}{k}(\eta) = \eta_i \underset{\sim}{b}^i$. We observe that η_i are not integers unless $\underset{\sim}{k}(\eta)$ is also a reciprocal lattice vector $\underset{\sim}{Y}$ so that $\eta_i = y_i$. Since $\underset{\sim}{k}$ is restricted to a reciprocal lattice cell η_i, which can be taken to be always positive, can be utmost unity. The boundary condition (13) now implies

$$\eta_i = \frac{h_i}{N_i} ; \quad h_i = 1,2,\ldots,N_i \quad . \tag{V.14}$$

Clearly there are N_i allowed values of η_i , the $N = \prod_{i=1}^{3} N_i$ values of $\underset{\sim}{k}$ will be called the N permitted wave numbers. Therefore it is seen that the assumed solution (7) satisfies both the uniqueness requirement and the boundary condition through a single condition (14) on $\underset{\sim}{k}$.

It is imperative that the number of modes of vibration in a finite crystal be equal to the number of degrees of freedom. The crystal which we have just considered contains N lattice cells with n particles in each cell, then the number of degrees of freedom is $3nN$. On the other hand, in the model there are N permitted values of $\underset{\sim}{k}$ and for each $\underset{\sim}{k}$ the secular determinant (9) yields $3n$ frequencies so the number of vibration modes is also $3nN$. Furthermore we note that the volume of the reciprocal lattice cell is just

$$|\underset{\sim}{b}^1 \cdot \underset{\sim}{b}^2 \times \underset{\sim}{b}^3| = \frac{1}{|\underset{\sim}{a}_1 \cdot \underset{\sim}{a}_2 \times \underset{\sim}{a}_3|} = \frac{1}{v_a} ,$$

where v_a is the volume of the lattice cell. This indicates that the permitted wave numbers $\underset{\sim}{k}$ are uniformly distributed in the reciprocal lattice with a density equal to $Nv_a = V$, the volume of the crystal.

VI. NORMAL COORDINATES+

The discussion in the previous section shows that the use of harmonic approximation and the periodic boundary condition leads to a Hamiltonian for a finite crystal of the form

$$H_s = \frac{1}{2}\sum_{\ell}^{N}\sum_{s\alpha}\left\{(M^s)^{-1}(T^s_{\ell\alpha})^2 + \sum_{\ell'}^{N}\sum_{s'\alpha'} U^{ss'}_{\ell-\ell'\alpha\alpha'} u^s_{\ell\alpha} u^{s'}_{\ell'\alpha'}\right\} , \qquad \text{(VI.1)}$$

where the first term is the usual kinetic-energy term with $T^s_{\ell\alpha} = \frac{\hbar}{i}\frac{\partial}{\partial u^s_{\ell\alpha}}$. Since the particle displacements in the potential are coupled, it becomes desirable to seek a new set of independent coordinates in terms of which the Hamiltonian appears as a sum of terms, each involving only one of these coordinates. When these coordinates are properly chosen, the transformed Hamiltonian will exhibit the same form as that for 3nN independent oscillators and any subsequent analysis will be considerably simplified. Such a set of coordinates is known as the set of normal coordinates.

It will be convenient to eliminate the presence of the particle mass by introducing reduced quantities. Thus

$$w^s_{\ell\alpha} = (M^s)^{1/2} u^s_{\ell\alpha} , \qquad \text{(VI.2)}$$

$$D^{ss'}_{\ell-\ell'\alpha\alpha'} = (M^s M^{s'})^{-1/2} U^{ss'}_{\ell-\ell'\alpha\alpha'} , \qquad \text{(VI.3)}$$

$$H_s = \frac{1}{2}\sum_{\ell s\alpha}\left\{(p^s_{\ell\alpha})^2 + \sum_{\ell' s'\alpha'} D^{ss'}_{\ell-\ell'\alpha\alpha'} w^s_{\ell\alpha} w^{s'}_{\ell'\alpha'}\right\} , \qquad \text{(VI-4)}$$

+ Part of this section follows the treatment of Born and Huang, Reference 8.

where $\quad p^s_{\ell\alpha} = \frac{\hbar}{i}\frac{\partial}{\partial w^s_{\ell\alpha}}$. Before actually introducing the normal coordinate transformations we will first examine certain useful properties of the solution (V.7). Consider the function

$$\begin{aligned}\Delta(\underset{\sim}{k}) &= \frac{1}{N}\sum_{\ell}^{N} e^{2\pi i \underset{\sim}{k}(\eta)\cdot\underset{\sim}{x}_\ell} \\ &= \frac{1}{N}\prod_{i=1}^{3}\frac{1-e^{2\pi i\eta_i N_i}}{1-e^{2\pi i\eta_i}} \quad .\end{aligned} \tag{VI.5}$$

Since $N_i\eta_i = h_i$ are integers, $\Delta(\underset{\sim}{k})$ vanishes everywhere except at the reciprocal lattice points where the denominators also vanish. That is to say, η_i will only be integers when $\underset{\sim}{k}(\eta)$ becomes a reciprocal lattice vector. Therefore,

$$\Delta(\underset{\sim}{k}) = \begin{cases} 1 & \text{all } \eta_i \text{ are integers} \\ 0 & \eta_i \text{ not all integers} \end{cases} \quad . \tag{VI.6}$$

Now construct the function

$$\Delta[-\underset{\sim}{k}(\eta')+\underset{\sim}{k}(\eta)] = \sum_{\ell}^{N}[(N)^{-1/2}e^{2\pi i\underset{\sim}{k}(\eta')\cdot\underset{\sim}{x}_\ell}]^*[(N)^{-1/2}e^{2\pi i\underset{\sim}{k}(\eta)\cdot\underset{\sim}{x}_\ell}] \ ,$$

which, according to (6), will vanish unless the argument of Δ is a reciprocal lattice vector. With the restriction on $\underset{\sim}{k}$ given by (V.14) the non-vanishing of Δ requires

$$-\underset{\sim}{k}(\eta') + \underset{\sim}{k}(\eta) = \underset{\sim}{k}(\eta-\eta') = \underset{\sim}{k}(o) \ .$$

Hence we obtain the orthonormal property

$$\frac{1}{N}\sum_{\ell}^{N}(e^{2\pi i\underset{\sim}{k}'\cdot\underset{\sim}{x}_\ell})^*(e^{2\pi i\underset{\sim}{k}\cdot\underset{\sim}{x}_\ell}) = \delta_{\underset{\sim}{k}\underset{\sim}{k}'} \quad , \tag{VI.7}$$

where $\underset{\sim}{k}$ and $\underset{\sim}{k}'$ denote two permitted wave numbers. By a similar argument the closure property of (10) is obtained,

$$\frac{1}{N}\sum_{\underset{\sim}{k}}^{N}(e^{2\pi i\underset{\sim}{k}\cdot\underset{\sim}{x}_{\ell'}})^{*}(e^{2\pi i\underset{\sim}{k}\cdot\underset{\sim}{x}_{\ell}}) = \delta_{\ell\ell'} \quad , \tag{VI.8}$$

where ℓ and ℓ' are two lattice cell indices.

The transformation to normal coordinates can now be derived. First expand the reduced displacements

$$w^{s}_{\ell\alpha} = (N)^{-1/2}\sum_{\underset{\sim}{k}}^{N} w^{s}_{\underset{\sim}{k}\alpha}e^{2\pi i\underset{\sim}{k}\cdot\underset{\sim}{x}_{\ell}} \ , \tag{VI.9}$$

with inverse

$$w^{s}_{\underset{\sim}{k}\alpha} = (N)^{-1/2}\sum_{\ell}^{N} w^{s}_{\ell\alpha}e^{-2\pi i\underset{\sim}{k}\cdot\underset{\sim}{x}_{\ell}} \ . \tag{VI.10}$$

The fact that the displacements must be real implies

$$w^{s}_{\underset{\sim}{k}\alpha} = w^{s}_{-\underset{\sim}{k}\alpha} \quad . \tag{VI.11}$$

As a result of (11), the potential part of (4) becomes

$$\frac{1}{2N}\sum_{\substack{\ell\ell' \\ \underset{\sim}{k}\underset{\sim}{k}'}}^{N}\sum_{\substack{s\alpha \\ s'\alpha'}} D^{ss'}_{\ell-\ell'\alpha\alpha'}w^{s}_{\underset{\sim}{k}\alpha}w^{s'}_{\underset{\sim}{k}'\alpha'}e^{2\pi i(\underset{\sim}{k}+\underset{\sim}{k}')\cdot\underset{\sim}{x}_{\ell}}$$

$$= \frac{1}{2}\sum_{\substack{s\alpha \\ s'\alpha' \\ \underset{\sim}{k}}} D^{ss'}_{\underset{\sim}{k}\alpha\alpha'}w^{s}_{\underset{\sim}{k}\alpha}w^{s'}_{-\underset{\sim}{k}\alpha'} \ ,$$

where

$$D^{ss'}_{\underset{\sim}{k}\alpha\alpha'} = \sum_{\ell} D^{ss'}_{\ell\alpha\alpha'}e^{-2\pi i\underset{\sim}{k}\cdot\underset{\sim}{x}_{\ell}} \quad .$$

Physically, expansion (9) describes the real displacement in terms of a series of N traveling waves with complex coefficients as amplitudes.

This unitary transformation reduces the potential to a series of N complex forms, each characterized by a $3n \times 3n$ matrix $D^{ss'}_{\underset{\sim}{k}\alpha\alpha'}$ which is Hermitian.+ For a given $\underset{\sim}{k}$ there exist $3n$ sets of eigenvectors $e^{s}_{\underset{\sim}{k}j\alpha}$ and eigenvalues $\omega^2_{\underset{\sim}{k}j}$, $j = 1,2,\ldots,3n$, satisfying

$$\omega^2_{\underset{\sim}{k}j} e^{s}_{\underset{\sim}{k}j\alpha} = \sum_{s'\alpha'} D^{ss'}_{\underset{\sim}{k}\alpha\alpha'} e^{s'}_{\underset{\sim}{k}j\alpha'} \quad , \tag{VI.12}$$

where the eigenvectors form a complete and orthonormal set,

$$\sum_{s\alpha} e^{s*}_{\underset{\sim}{k}j\alpha} e^{s}_{\underset{\sim}{k}j'\alpha} = \delta_{jj'} \quad ,$$

$$\sum_{j} e^{s}_{\underset{\sim}{k}j\alpha} e^{s'}_{\underset{\sim}{k}j\alpha'} = \delta_{\alpha\alpha'}\delta_{ss'} \quad . \tag{VI.13}$$

Furthermore,+

$$e^{s*}_{\underset{\sim}{k}j}(\alpha) = e^{s}_{-\underset{\sim}{k}j}(\alpha) \quad . \tag{VI.14}$$

It is interesting to note the similarity between Equations (V.8) and (12). In fact $D^{ss'}_{\underset{\sim}{k}\alpha\alpha'}$ is completely equivalent to the matrix $C^{ss'}_{\underset{\sim}{k}\alpha\alpha'}$ except for a difference in the phase factor. Accordingly, the corresponding eigenvectors g^{s}_{α}, more explicitly written as $g^{s}_{\underset{\sim}{k}j\alpha}$, differ from $e^{s}_{\underset{\sim}{k}j\alpha}$ by a compensating phase factor $\exp(-2\pi i\underset{\sim}{k}\cdot\underset{\sim}{x}^{s})$. Since the two eigenvalues are the same for a given $\underset{\sim}{k}$, the choice of phase factors is arbitrary.

As noted earlier the need for normal coordinates arises from the fact that in Equation (1) the particle displacements are dynamically coupled through the three indices referring to lattice cell, position in the cell, and direction of displacement. Since the normal coordinates will not be coupled in the Hamiltonian, they must not depend on ℓ, s, and α. The

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+ Proof given in Appendix A.

transformation (11) has eliminated the coupling through cell index ℓ by introducing the dependence on $\underset{\sim}{k}$. It follows that the next step should be one which will transform the dependence on s, α to a dependence on other indices. This can be achieved by using the eigenvectors just found. So we write

$$w^{s}_{\underset{\sim}{k}\alpha} = \sum_{i} e^{s}_{\underset{\sim}{k}j\alpha} Q_{\underset{\sim}{k}j} , \tag{VI.15}$$

with inverse by virtue of (13)

$$Q_{\underset{\sim}{k}j} = \sum_{s\alpha} w^{s}_{\underset{\sim}{k}\alpha} e^{s*}_{\underset{\sim}{k}j\alpha} . \tag{VI.16}$$

On account of the fact that (s,α) in the dynamical matrix $D^{ss'}_{\underset{\sim}{k}\alpha\alpha'}$ can be taken as a single index, only a single new index j, $j=1,2,\ldots,3n$, is required in the transformation.

Now the Hamiltonian (1), when expressed in terms of the coordinates $Q_{\underset{\sim}{k}j}$, takes the form

$$H_s = \frac{1}{2} \sum_{\underset{\sim}{k}}^{N} \sum_{j=1}^{3n} \left\{ \left(\frac{\hbar}{i}\right)^2 \left(\frac{\partial}{\partial Q_{\underset{\sim}{k}j}}\right)^* \left(\frac{\partial}{\partial Q_{\underset{\sim}{k}j}}\right) \right.$$

$$\left. + \omega^2_{\underset{\sim}{k}j} Q^*_{\underset{\sim}{k}j} Q_{\underset{\sim}{k}j} \right\} , \tag{IV.17}$$

where

$$Q^*_{\underset{\sim}{k}j} = \sum_{s\alpha} e^{s}_{\underset{\sim}{k}j\alpha} w^{s*}_{\underset{\sim}{k}\alpha} = \sum_{s\alpha} e^{s*}_{-\underset{\sim}{k}j\alpha} w^{s}_{-\underset{\sim}{k}\alpha} = Q_{-\underset{\sim}{k}j} .$$

We will regard the set of coordinates $Q_{\underset{\sim}{k}j}$ as the $3nN$ complex normal coordinates related to the actual particle coordinate by the expression

$$u^{s}_{\ell\alpha} = (M^s N)^{-1/2} \sum_{\underset{\sim}{k}}^{N} \sum_{j}^{3n} Q_{\underset{\sim}{k}j} e^{s}_{\underset{\sim}{k}j\alpha} e^{2\pi i \underset{\sim}{k}\cdot \underset{\sim}{x}_{\ell}} . \tag{VI.18}$$

In the study of neutron interaction with crystal two different kinds of normal coordinates can be used. We will next show how they may be derived from the coordinates $Q_{\underset{\sim}{k}j}$ just obtained. It will be seen that the first kind of normal coordinate corresponds to a combination of the complex waves described by $Q_{\underset{\sim}{k}j}$ and its complex conjugate $Q^*_{\underset{\sim}{k}j}$ to give two real standing waves while the second kind of normal coordinates involves a reformulation of the problem in which the coordinate and its canonical conjugate momentum are expressed in terms of quantum mechanical operators known as "creation" and "annihilation" operators. Physically, the latter type of coordinates is related to traveling waves moving in opposite directions.[9]

A. Standing Waves

Observe that for $\underset{\sim}{k} = 0$, $Q^*_{oj} = Q_{oj}$ so that these coordinates can be used directly as real normal coordinates. For $\underset{\sim}{k} \neq 0$ the permitted wave numbers can be divided into two groups by passing an arbitrary plane through the origin in reciprocal space. Therefore, to every allowable $\underset{\sim}{k}$ on the "positive" side there corresponds a $-\underset{\sim}{k}$ on the opposite side of the plane. Since the Hamiltonian is symmetric in $\underset{\sim}{k}$ it is then only necessary to sum over half the reciprocal cell. Let $Q_{\underset{\sim}{k}j} = \frac{1}{\sqrt{2}}(z_1 + iz_2)$, where z_i, $i = 1,2$, are two real, independent coordinates and we have suppressed the indices $\underset{\sim}{k}$ and j. Under this additional transformation the Hamiltonian (17) becomes

$$H_s = \frac{1}{2} \sum_{\underset{\sim}{k}}^{N/2} \sum_{j=1}^{3n} \sum_{i=1}^{2} \{p_i^2 + \omega^2_{\underset{\sim}{k}j} z_i^2\} ,$$

with $p_i = \frac{\hbar}{i}\frac{\partial}{\partial z_i}$, which is the familiar form of a system of $3nN$ uncoupled harmonic oscillators. Each of the $3N/2$ frequencies is shared by a z_1 oscillator and a z_2 oscillator. In terms of these real normal coordinates the particle displacement has the form

$$u^s_{\ell\alpha} = (2M^sN)^{-1/2} \sum_{\underset{\sim}{k}j} \{z_1(e^s_{\underset{\sim}{k}j\alpha}e^{2\pi i\underset{\sim}{k}\cdot\underset{\sim}{x}_\ell} + e^{s*}_{\underset{\sim}{k}j\alpha}e^{-2\pi i\underset{\sim}{k}\cdot\underset{\sim}{x}_\ell})$$

$$+ iz_2(e^s_{\underset{\sim}{k}j\alpha}e^{2\pi i\underset{\sim}{k}\cdot\underset{\sim}{x}_\ell} + e^{s*}_{\underset{\sim}{k}j\alpha}e^{-2\pi i\underset{\sim}{k}\cdot\underset{\sim}{x}_\ell})\} \quad .$$

In the special case of monatomic crystal, the particle displacement can be written as

$$u_{\ell\alpha} = (\frac{2}{MN})^{1/2} \sum_{\underset{\sim}{k}j} A_{\underset{\sim}{k}j\alpha} \{z_1 \cos(\underset{\sim}{k}\cdot\underset{\sim}{x}_\ell) + z_2 \sin(\underset{\sim}{k}\cdot\underset{\sim}{x}_\ell)\} \ ,$$

where $A_{\underset{\sim}{k}j\alpha}$ is the α-th component of the phonon polarization vector $\underset{\sim}{A}_{\underset{\sim}{k}j}$. The normal coordinates in this form have been used in the early investigation of neutron scattering by crystals.(1)

B. "Creation" and "Annihilation" Operators

Consider the Hamiltonian given by (17),

$$H = \frac{1}{2} \sum_{\lambda} (p^*_\lambda p_\lambda + \omega_\lambda Q^*_\lambda Q_\lambda) \ , \qquad \text{(VI.18)}$$

where the indices $\underset{\sim}{k}$ and j have been replaced by λ for convenience and $p_\lambda = \frac{\hbar}{i}\frac{\partial}{\partial Q_\lambda}$. Coordinate Q_λ and conjugate momentum p_λ are operators which satisfy the commutation rules,

$$[Q_\lambda, p_{\lambda'}] = i\hbar\,\delta_{\lambda\lambda'}$$

$$[Q_\lambda, Q^*_\lambda] = [p_\lambda, p^*_\lambda] = [Q_\lambda, p^*_\lambda] = 0 \ . \qquad \text{(VI.19)}$$

In order to examine the time dependence of Q_λ and p_λ, we next consider the equations of motion,[10]

$$i\hbar \frac{dQ_\lambda}{dt} = [Q_\lambda, H] = i\hbar\, p_\lambda^* \quad ,$$

$$i\hbar \frac{dp_\lambda}{dt} = [p_\lambda, H] = -i\hbar\omega^2 Q_\lambda^* \ ,$$

where we have used the fact that summation over $\underset{\sim}{k}$ includes both positive and negative wave numbers in evaluating the commutators. The equations are then easily solved to give

$$Q_\lambda = a_\lambda e^{-i\omega_\lambda t} + b_\lambda e^{i\omega_\lambda t} \quad ,$$

with $p_\lambda = \frac{dQ_\lambda^*}{dt}$. Corresponding expressions exist for Q_λ^* and p_λ^* . Since $Q_\lambda^* = Q_{-\lambda}$, where the sign of λ corresponds to that of $\underset{\sim}{k}$ while index j is always positive, the relation between the time-independent operators a_λ, b_λ is

$$a_\lambda^* = b_{-\lambda} \ .$$

These operators can be written in terms of Q_λ and p_λ ,

$$a_\lambda = \frac{1}{2}(Q_\lambda + \frac{i}{\omega_\lambda} p_\lambda^*), \qquad b_\lambda = \frac{1}{2}(Q_\lambda - \frac{i}{\omega_\lambda} p_\lambda^*) \ ,$$

and they satisfy the commutation relation

$$[a_\lambda, a_{\lambda'}^*] = \frac{\hbar}{2\omega_\lambda}\, \delta_{\lambda\lambda'} \ .$$

We can therefore define new operators, $\zeta_\lambda = (\frac{2\omega_\lambda}{\hbar})^{1/2} a_\lambda e^{-i\omega_\lambda t}$ so that the Hamiltonian (18) becomes

$$H = \hbar \sum_\lambda \omega_\lambda (\zeta_\lambda^* \zeta_\lambda + \frac{1}{2}) \ , \qquad \text{(VI.20)}$$

with

$$[\zeta_\lambda, \zeta^*_{\lambda'}] = \delta_{\lambda\lambda'} \; . \tag{VI.21}$$

The original particle displacement is now expressed as

$$\begin{aligned} u_{\ell\alpha}(t) &= \sum_\lambda e^s_{\lambda\alpha} e^{2\pi i \underset{\sim}{k}\cdot\underset{\sim}{x}_\ell} Q_\lambda(t) \\ &= \left(\frac{\hbar}{2M^s N}\right)^{1/2} \sum_\lambda \omega_\lambda^{-1/2} \{ \zeta_\lambda e^s_{\lambda\alpha} e^{2\pi i \underset{\sim}{k}\cdot\underset{\sim}{x}_\ell} \\ &\quad + \zeta^*_\lambda e^s_{\lambda\alpha} e^{-2\pi i k\cdot x_\ell} \} \; . \end{aligned} \tag{VI.22}$$

In arriving at (20) and (22) one must keep in mind that λ can be positive and negative because of the summation over $\underset{\sim}{k}$. Equation (22) is commonly used to describe particle motions in studies of neutron interaction with crystals.[3] For the Einstein crystal the particle displacements are the normal coordinates for the system, and upon transformation become

$$u^s_{\ell\alpha}(t) = \left(\frac{\hbar}{2M^s\omega}\right)(\zeta^s_\ell + \zeta^{s*}_\ell) \; , \tag{VI.22a}$$

where it is to be expected that $u^s_{\ell\alpha}(t)$ is independent of α.

We proceed to show that ζ^*_λ and ζ_λ operating on the wave function of the crystal will result in the emission and absorption of a quantum of sound radiation (energy $\hbar\omega_\lambda$), known as a phonon, and are therefore the "creation" and "annihilation" operators. It is noted that (18) is in the form of a Hamiltonian for $3nN$ uncoupled harmonic oscillators, therefore the energy of the λ-th oscillator in the n-th eigenstate is

$$\langle n_\lambda | H_\lambda | n_\lambda \rangle = (n_\lambda + \tfrac{1}{2})\hbar\,\omega_\lambda \; , \tag{VI.23}$$

where

$$H_\lambda = \frac{1}{2}\,(p^*_\lambda p_\lambda + \omega^2_\lambda Q^*_\lambda Q_\lambda) \; ,$$

and $|n_\lambda>$ denotes the wave function of the λ-th oscillator in the n-th eigenstate. n_λ is the quantum number of the oscillator and can also be regarded as the number of λ-phonons in the system. According to (20) and (21), we have

$$(\hbar\omega_\lambda)^{-1}(H_\lambda + \tfrac{1}{2}) = \zeta_\lambda\zeta_\lambda^* \ ; \quad (\hbar\omega_\lambda)^{-1}(H_\lambda - \tfrac{1}{2}) = \zeta_\lambda^*\zeta_\lambda \ . \qquad \text{(VI.24)}$$

Operate the first equation on ζ_λ,

$$(\hbar\omega_\lambda)^{-1}(H_\lambda + \tfrac{1}{2})\zeta_\lambda = \zeta_\lambda\zeta_\lambda^*\zeta_\lambda = (\hbar\omega_\lambda)^{-1}\ \zeta_\lambda(H_\lambda - \tfrac{1}{2})$$

so that

$$H_\lambda\zeta_\lambda|n_\lambda> = (n_\lambda-1)\zeta_\lambda|n_\lambda> \ . \qquad \text{(VI.25)}$$

Equation (25) shows that $\zeta_\lambda|n_\lambda>$ is proportional to an eigenfunction of H_λ. Let the proportionality constant be A_n, a constant depending upon n, then we can write

$$\zeta_\lambda|n_\lambda> = A_n|n_\lambda-1> \ .$$

Similarly it can be shown that

$$\zeta_\lambda^*|n_\lambda> = B_n|n_\lambda+1> \ ,$$

where B_n is another constant depending upon n.

To determine A_n and B_n we simply observe that

$$<n_\lambda|\zeta_\lambda\zeta_\lambda^*|n_\lambda> = B_n^2 \ ; \quad <n_\lambda|\zeta_\lambda^*\zeta_\lambda|n_\lambda> = A_n^2 \ .$$

Comparing the Equations (23) and (24) it is seen that $A_n = \sqrt{n_\lambda}$ and $B_n = \sqrt{n_\lambda+1}$.

VII. SCATTERING OF NEUTRONS BY A MONATOMIC CRYSTAL

Scattering by a monatomic crystal is perhaps the simplest case in which all the essental aspects of the dependence of cross section upon the structure and dynamical behavior of the scattering system can be investigated. The results derived in this section can be generalized in a straightforward manner to the polyatomic case by the use of structure factor. We now use the crystal model which has just been developed but will simplify it to the case of only one particle occupying each cell. The differential cross section will be calculated using the formalism developed earlier and interpretations can then be made which are basic to a general understanding of neutron interactions with crystals.

We first consider the factor in Equation (III.11),

$$\langle \chi_{\ell\ell'} \rangle_T = \sum_n P_n \langle n | \chi_{\ell\ell'} | n \rangle \ , \tag{VII.1}$$

with

$$\chi_{\ell\ell'} = e^{i\underset{\sim}{\kappa}\cdot\underset{\sim}{R}_{\ell'}(t)} e^{-i\underset{\sim}{\kappa}\cdot\underset{\sim}{R}_{\ell}} \ . \tag{VII.2}$$

Here $\langle\ \rangle_T$ is used to denote the average over all possible initial states n (previously n_o) of the crystal. The crystal is taken to be in thermodynamic equilibrium with the surroundings so this average is often called the thermal average. In terms of the normal mode analysis we can approximately describe the dynamical behavior of the N-particle crystal by that of a system of 3N independent harmonic oscillators. It will be seen that use of the "creation" and "annihilation" operators greatly simplifies the calculation and that the explicit form of $|n\rangle$ is never needed. The instantaneous position of the particle in ℓ-th cell can be written as

$$\underset{\sim}{R}_\ell = \underset{\sim}{x}_\ell + \sum_\lambda \underset{\sim}{c}_{\ell\lambda} r_\lambda \ , \qquad \text{(VII.3)}$$

where the decomposition into normal coordinates r_λ is written formally. Whenever explicit forms are required, r_λ are the "annihilation" and "creation" operators, i.e.,

$$\sum_\lambda \underset{\sim}{c}_{\ell\lambda} r_\lambda = \sum_\lambda (\underset{\sim}{Z}_{\ell\lambda}\zeta_\lambda + \underset{\sim}{Z}^*_{\ell\lambda}\zeta^*_\lambda) \ , \qquad \text{(VII.4)}$$

with

$$\underset{\sim}{Z}_{\ell\lambda} = \left(\frac{\hbar}{2MN\omega_\lambda}\right)^{1/2} \underset{\sim}{A}_\lambda e^{2\pi i \underset{\sim}{k}\cdot \underset{\sim}{x}_\ell} \ ,$$

where it is understood that index λ represents one of the N permitted wave numbers and one of the three directions of polarization. Thus $A_\lambda = \underset{\sim}{A}_j(\underset{\sim}{k})$ is the j-th unit polarization vector appropriate to that oscillator whose propagation vector is $\underset{\sim}{k}$. We note that in the quantum formalism $\underset{\sim}{x}_\ell$, the equilibrium position of the ℓ-th particle, is not a dynamical variable and that the normal coordinates can appear as time-independent operators r_λ (or ζ_λ) in the Schrödinger picture, or as time-dependent operators $r(t)$ [or $\zeta_\lambda(t)$] in the Heisenberg picture. As we have shown, the difference between ζ_λ and $\zeta_\lambda(t)$ is the time factor $\exp(-i\omega_\lambda t)$. The commutation rule and eigenvalues of these operators have been given earlier.

The operator (2) now takes the form

$$\chi_{\ell\ell'} = e^{i\underset{\sim}{\kappa}\cdot(\underset{\sim}{x}_{\ell'} - \underset{\sim}{x}_\ell)} \, e^{i\underset{\sim}{\kappa}\cdot\sum_\lambda Z_{\ell'\lambda} r_\lambda(t)} \, e^{-i\kappa\cdot\sum Z_{\ell\lambda} r_\lambda}$$

$$= e^{i\underset{\sim}{\kappa}\cdot(x_{\ell'} - x_\ell)} \prod_\lambda e^{i\kappa\cdot Z_{\ell'\lambda} r_\lambda(t)} \, e^{-i\kappa\cdot Z_{\ell\lambda} r_\lambda} \ ,$$

since the normal coordinates are independent and therefore commute. Next we observe that for any two operators A and B which commute with their commutator [A,B] there exists the identity[+]

$$e^A e^B = e^{A+B+\frac{1}{2}[A,B]} \quad .$$

We make use of this fact to write

$$X_{\ell\ell'} = e^{i\kappa\cdot(x_{\ell'}-x_{\ell})} \prod_{\lambda} e^{\frac{1}{2}(\kappa\cdot Z_{\ell'\lambda})(\kappa\cdot Z_{\ell\lambda})[r_\lambda(t),r_\lambda]} e^{i\kappa\cdot\{Z_{\ell'\lambda}r_\lambda(t) - Z_{\ell\lambda}r_\lambda\}}$$

The thermal average (1) becomes

$$\begin{aligned} \langle X_{\ell\ell'}\rangle_T &= e^{i\kappa\cdot(x_{\ell'}-x_{\ell})} \sum_n \prod_\lambda P_{n_\lambda}\langle n_\lambda|X^\lambda_{\ell\ell'}|n_\lambda\rangle \\ &= e^{i\kappa\cdot(x_{\ell'}-x_{\ell})} \prod_\lambda \langle X^\lambda_{\ell\ell'}\rangle_T, \end{aligned} \quad \text{(VII.5)}$$

where the initial crystal eigenfunction $|n\rangle$ has been written explicity as a product of oscillator eigenfunctions $|n_\lambda\rangle$, $\lambda = 1,2,\ldots,3N$, and

$$\begin{aligned} \langle X^\lambda_{\ell\ell'}\rangle_T &= \sum_{n_\lambda} P_{n_\lambda} \langle n_\lambda|X^\lambda_{\ell\ell'}|n_\lambda\rangle \\ &= e^{\frac{1}{2}(\kappa\cdot Z_{\ell'\lambda})(\kappa\cdot Z_{\ell\lambda})[r_\lambda(t),r_\lambda]} \langle e^{i\kappa\cdot\{Z_{\ell'\lambda}r_\lambda(t)-Z_{\ell\lambda}r_\lambda\}}\rangle_T , \end{aligned} \quad \text{(VII.6)}$$

the commutator here being just a number. It is noted that we have written P_n, the probability of finding the crystal initially in state n, as a product of P_{n_λ}'s , where P_{n_λ} is the probability of finding the λ-th oscillator initially in state n. Again this is a direct consequence of the normal mode analysis and the fact that the crystal is in thermodynamic

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[+] Proof given in Appendix B.

equilibrium. The density matrix is diagonal and is explicitly

$$P_{n_\lambda} = e^{-2n\nu_\lambda}\left(\sum_{n_\lambda} e^{-2n\nu_\lambda}\right)^{-1}$$

$$= e^{-2n\nu_\lambda}(1 - e^{-2\nu_\lambda}), \tag{VII.7}$$

where ν_λ has been defined in Section IV. It is seen that phonons obey Bose-Einstein statistics.

In order to calculate the thermal average of $X^\lambda_{\ell\ell'}$ we make use of the corollary to Block's theorem[4]

$$\langle e^Q \rangle_T = e^{\frac{1}{2}\langle Q^2 \rangle_T} , \tag{VII.8}$$

where Q is a multiple of, or some linear combination of, commuting oscillator coordinates and their conjugate momenta. Then

$$\langle X^\lambda_{\ell\ell'} \rangle_T = \exp\{-\tfrac{1}{2}[(\underset{\sim}{\kappa}\cdot\underset{\sim}{Z}_{\ell'\lambda})^2\langle r^2_\lambda(t)\rangle_T + (\underset{\sim}{\kappa}\cdot\underset{\sim}{Z}_{\ell\lambda})^2\langle r^2_\lambda\rangle_T$$

$$- 2(\kappa\cdot Z_{\ell'\lambda})(\kappa\cdot Z_{\ell\lambda})\langle r_\lambda(t), r_\lambda\rangle_T]\} . \tag{VII.9}$$

The thermal average of the time-dependent and time-independent normal coordinates can be readily obtained,

$$\langle r^2_\lambda(t)\rangle_T = \langle r^2_\lambda\rangle_T = 2\langle n_\lambda\rangle_T + 1 ,$$

$$\langle r_\lambda(t) r_\lambda\rangle_T = e^{-i\omega_\lambda t}\langle n_\lambda + 1\rangle + e^{i\omega_\lambda t}\langle n_\lambda\rangle_T ,$$

where

$$\langle n_\lambda\rangle_T = \sum_{n_\lambda} n_\lambda P_{n_\lambda} = e^{-2\nu_\lambda}(1-e^{-2\nu_\lambda})^{-1} .$$

Equation (9) becomes

$$\langle\alpha_{\ell\ell'}^{\lambda}\rangle_T = e^{-D_\lambda(\underset{\sim}{\kappa}\cdot\underset{\sim}{A}_\lambda)^2} \sum_{n=-\infty}^{\infty} e^{-n\nu_\lambda} e^{-in\{2\pi\underset{\sim}{k}\cdot(\underset{\sim}{x}_{\ell'}-\underset{\sim}{x}_\ell)-\omega_\lambda t\}}$$

$$(x)\ I_n(P_\lambda(\underset{\sim}{\kappa}\cdot\underset{\sim}{A}_\lambda)^2) \quad , \qquad \text{(VII.10)}$$

where

$$D_\lambda = \frac{\hbar \coth \nu_\lambda}{2MN\omega_\lambda} \ , \quad P_\lambda = \frac{\hbar \operatorname{csch} \nu_\lambda}{2MN\omega_\lambda}$$

and we have rearranged the result by using the generating function of the modified Bessel function of first kind,

$$e^{\frac{1}{2} r(t + \frac{1}{t})} = \sum_{n=-\infty}^{\infty} t^n I_n(r) \quad .$$

We can now exhibit the energy- and angle-dependent differential scattering cross section for a monatomic single crystal of N spinless particles as

$$\sigma(\rho,\underset{\sim}{\Omega}) = \frac{k_f}{2\pi N k_i \hbar} \int dt e^{-i\rho t} \sum_{\ell\ell'} a_\ell a_{\ell'} e^{i\underset{\sim}{\kappa}\cdot(\underset{\sim}{x}_{\ell'}-\underset{\sim}{x}_\ell)} \prod_\lambda \langle\alpha_{\ell\ell'}^{\lambda}\rangle_T$$

$$= \frac{k_f e^{-2W}}{2\pi k_i \hbar} \int dt e^{-i\rho t} \prod_\lambda \sum_{m_\lambda=-\infty}^{\infty} e^{-m_\lambda \nu_\lambda} I_{m_\lambda}(P_\lambda(\underset{\sim}{\kappa}\cdot\underset{\sim}{A}_\lambda)^2)$$

$$(x)\ F_{m_\lambda} e^{i m_\lambda \omega_\lambda t} \quad , \qquad \text{(VII.11)}$$

where

$$\exp(-2W) = \exp\{-\sum_\lambda D_\lambda(\underset{\sim}{\kappa}\cdot\underset{\sim}{A}_\lambda)^2\} \ ,$$

$$F_{m_\lambda} = \frac{1}{N} \sum_{\ell\ell'} a_\ell a_{\ell'} \exp\{i(\underset{\sim}{\kappa}-2\pi m_\lambda \underset{\sim}{k})\cdot(\underset{\sim}{x}_{\ell'}-\underset{\sim}{x}_\ell)\} \ .$$

Since the present crystal model assumes harmonic vibrations of the particles then it should be possible to reduce the above result to that for the

Einstein crystal. The necessary specialization required is indicated from a comparison of Equations (IV.22) and (IV.22a). Hence we obtain for the monatomic case,

$$<X_{\ell\ell}>_T = e^{-D\kappa^2} \sum_{n=-\infty}^{\infty} e^{-n\nu} e^{-in\omega t} I_n(P\kappa^2) ,$$

where D, P, ν have been defined in Section IV. Note that the reduction applies only for $\ell'=\ell$. For $\ell'\neq\ell$ the thermal average is time-independent because

$$<X_{\ell'\ell}>_T = \sum P_n <n|\Lambda_{\ell'}^{+}(t)\Lambda_\ell|n>$$

$$= <\Lambda_{\ell'}^{+}>_T \; <\Lambda_\ell>_T ,$$

where

$$<\Lambda_\ell>_T = \sum_{n_\ell} P_{n_\ell} <n_\ell|\Lambda_\ell|n_\ell> ,$$

and $|n_\ell>$ is the eigenstate of the particle in the ℓ-th cell. The corresponding differential scattering cross section for the Eienstein crystal is

$$\sigma(\rho,\underset{\sim}{\Omega}) = \frac{k_f e^{-D\kappa^2}}{2\pi N k_1 \hbar} \int dt e^{-i\rho t} \{\sum_\ell a_\ell^2 \sum_{n=-\infty}^{\infty} e^{-n(\nu+i\omega t)} I_n(P\kappa^2)$$

$$+ \sum_{\ell'\ell} a_{\ell'} a_\ell e^{i\underset{\sim}{\kappa}\cdot(\underset{\sim}{x}_{\ell'}-\underset{\sim}{x}_\ell)}\} . \qquad \text{(VII-11a)}$$

We have shown that a system of oscillators can undergo energy transitions only in discrete amounts; the λ-th oscillator can transfer an amount equal to integral multiple of its energy level spacing $\hbar\omega_\lambda$. Therefore for any neutron-crystal interaction the energy gain or loss of the neutron can be written as

$$\rho = \sum_\lambda n_\lambda \omega_\lambda , \qquad \text{(VII.12)}$$

where each integer n_λ can be positive or negative. Accordingly we perform the indicated time integration to obtain from Equation (11)

$$\sigma(\rho,\underset{\sim}{\Omega}) = \frac{k_f e^{-2W}}{k_i} \sum_{\{m_\lambda\}} \delta(\sum_\lambda \hbar(n_\lambda - m_\lambda)\omega_\lambda) \prod_\lambda F_{m_\lambda}$$

$$(x)\ I_{m_\lambda}(P_\lambda(\underset{\sim}{\kappa}\cdot\underset{\sim}{A}_\lambda)^2)e^{-m_\lambda\nu_\lambda} \quad . \qquad \text{(VII.13)}$$

The energy conservation conditions appears explicitly as the delta function. The indicated summation is over all possible sets of $\{m_\lambda\}$ and for a given set $\{m_\lambda\}$ each m_λ in the product is specified by the set according to λ.

In an energy measurement the effect of the finite resolution of the instrument is essentially to integrate the measured quantity over a small interval about the energy of the scattered neutrons. When treated in this manner the differential cross section $\sigma(\rho,\underset{\sim}{\Omega})$ is non-zero only for $\sum_\lambda (n_\lambda - m_\lambda)\omega_\lambda = 0$. For a given ρ we note that the set of integers $\{n_\lambda\}$ may not be unique. This is to say, there may exist different multi-oscillator excitation processes which can result in the same net energy exchange. We shall, however, ignore this aspect and assume, for the purpose of subsequent discussion, that each set $\{n_\lambda\}$ corresponds uniquely to a particular neutron energy transfer as written in (12). Thus it becomes possible to identify the coefficient of each delta function as the physically measured $\sigma(\rho,\underset{\sim}{\Omega})$, for example,

$$\sigma(\rho,\underset{\sim}{\Omega}) = \frac{k_f e^{-2W}}{k_i} \prod_\lambda F_{n_\lambda} e^{-n_\lambda\nu_\lambda} I_{n_\lambda}(P_\lambda(\underset{\sim}{\kappa}\cdot\underset{\sim}{A}_\lambda)^2). \qquad \text{(VII.14)}$$

In this way the elastic as well as all the inelastic cross sections can be derived from Equation (13).

We next examine the structure factor F_{n_λ}. It is convenient to remove it from the product by writing

$$F = \frac{1}{N} \sum_{\ell\ell'} a_\ell a_{\ell'} e^{i\underset{\sim}{\gamma}\cdot(\underset{\sim}{x}_{\ell'} - \underset{\sim}{x}_\ell)} , \qquad \text{(VII.15)}$$

where $\underset{\sim}{\gamma} = \underset{\sim}{\kappa} - \sum_\lambda n_\lambda \underset{\sim}{q}$, $\underset{\sim}{q} = 2\pi \underset{\sim}{k}$ and indices ℓ and ℓ' range over all lattice sites in the crystal. When the crystal contains two or more isotopes for which the scattering lengths differ the summation process effectively averages the scattering length over all the isotopes present. We can exhibit the double sum as two partial sums,

$$\sum_{\ell\ell'} a_\ell a_{\ell'} e^{i\underset{\sim}{\gamma}\cdot(\underset{\sim}{x}_{\ell'} - \underset{\sim}{x}_\ell)} = \sum_\ell a_\ell^2 + \sum_{\ell\ell'}{}' a_\ell a_{\ell'} e^{i\underset{\sim}{\gamma}\cdot(\underset{\sim}{x}_{\ell'} - \underset{\sim}{x}_\ell)} , \qquad \text{(VII.16)}$$

where the prime indicates that terms with $\ell = \ell'$ are to be omitted. The first sum in the separation can be interpreted as representing direct scattering effects while the second term involves scattering due to different particles and therefore represents interference effects. For a relatively simple model it will be instructive to discuss neutron interaction in terms of direct and interference scatterings and show the dependence of the two effects on different physical aspects of the scattering system. Essentially this was our approach in the discussion of the Einstein crystal. To carry out the indicated summation let C_A be the fractional concentration of isotope A which has scattering length a_A, etc. Then

$$\begin{aligned}
\sum_{\ell\ell'} a_\ell a_{\ell'} e^{i\underset{\sim}{\gamma}\cdot(\underset{\sim}{x}_{\ell'} - \underset{\sim}{x}_\ell)} &= (c_A a_A^2 + c_B a_B^2 + \dots) + (c_A a_A + c_B a_B + \dots)^2 \sum_{\ell\ell'}{}' e^{i\underset{\sim}{\gamma}\cdot(\underset{\sim}{x}_{\ell'} - \underset{\sim}{x}_\ell)} \\
&= N \langle a^2 \rangle + \langle a \rangle^2 \sum_{\ell\ell'}{}' e^{i\underset{\sim}{\gamma}\cdot(\underset{\sim}{x}_{\ell'} - \underset{\sim}{x}_\ell)} \\
&= N \left\{ \langle a^2 \rangle - \langle a \rangle^2 \right\} + \langle a \rangle^2 \left| \sum e^{i\underset{\sim}{\gamma}\cdot\underset{\sim}{x}_\ell} \right|^2 . \qquad \text{(VII.17)}
\end{aligned}$$

In the second sum we have assumed that the crystal is completely disordered. That is to say, the existence of an atom of a particular isotope at ℓ is independent of the type of atom at ℓ'. The symbol $< >$ is used to denote isotopic average. We will express γ in terms of reciprocal lattice basic vectors so that

$$\gamma \cdot x_\ell = \sum_{s=1}^{3} \gamma_s \ell^s \quad ,$$

where γ_s and ℓ^s are the respective components, then

$$\left| \sum_{\ell}^{N} e^{i\gamma \cdot x_\ell} \right|^2 = \prod_{s=1}^{3} \left| \sum_{\ell^s=1}^{N_s} e^{i\gamma_s \ell^s} \right|^2$$

$$= \prod_{s=1}^{3} \frac{\sin^2(N_s\gamma_s/2)}{\sin^2(\gamma_s/2)} \quad , \qquad \text{(VII.18)}$$

where N_s is the highest integer that ℓ^s can assume, i.e. $\prod_{s=1}^{3} N_s = N$. It is noted that this expression will be small compared to its value when the denominator vanishes. Since the denominator vanishes for $\gamma_s = 2\pi\mu_s$, $\mu_s = 0, \pm 1, \ldots$, we only need to examine the behavior of (18) in the neighborhood of these values of γ_s. Let $\gamma_s = 2\pi\mu_s + \epsilon$, ϵ small, then

$$\frac{\sin^2(N_s\gamma_s/2)}{\sin^2(\gamma_s/2)} \cong \frac{\sin^2(N_s\epsilon/2)}{(\epsilon/2)^2} \sim 2\pi N_s \delta(\epsilon) \ .$$

As a result we obtain

$$\left| \sum_{\ell}^{N} e^{i\gamma \cdot x_\ell} \right|^2 = (2\pi)^3 N \prod_{s=1}^{3} \delta(\gamma_s - 2\pi y_s) \ . \qquad \text{(VII.19)}$$

Equation (19) shows that the difference between momentum transfer vector κ and multiple phonon vector $\sum_\lambda n_\lambda q$ is a vector Y defined in reciprocal lattice space by

$$Y = \sum_{s=1}^{3} y_s b^s \quad ,$$

where, as we have seen in Section IV, a certain property of crystal structure is expressed through $\underset{\sim}{Y}$. Thus the influence of lattice geometry appears only in the above delta functions.[+] In elastic scattering these functions express the well-known Bragg condition originally proposed for X-ray interference effects.

The single crystal differential scattering cross section can now be written as

$$\sigma(\rho,\underset{\sim}{\Omega}) = \frac{k_f e^{-2W}}{k_i} \left\{a_I^2 + \frac{(2\pi)^3}{v_a} a_C^2 \delta(\underset{\sim}{\gamma}-2\pi\underset{\sim}{Y})\right\}$$

$$(x) \prod_{\lambda} e^{-n_\lambda \nu_\lambda} I_{n_\lambda}(P_\lambda(\underset{\sim}{\kappa}\cdot\underset{\sim}{A}_\lambda)^2) \quad . \tag{VII.20}$$

where $a_I^2 = \langle a^2\rangle - \langle a\rangle^2$, $a_C^2 = \langle a\rangle^2$, and the delta function is expressed in cartesian components.[++] The part of cross section containing a_I^2 is called incoherent while that containing a_C^2 is called coherent.[+++] We observe that the source of incoherence is the existence of isotopic mixture. If the crystal were monoisotopic then $a_I^2 = 0$. In the next section incoherence due to nuclear spin effects will be discussed.

[+] It is important to keep in mind the fact that use of the delta functions to represent mathematically a sharply peaked physical behavior is made for convenience.

[++] Since the delta function in question is defined with respect to a three-dimensional integral the Jacobian involved in the transformation is just the ratio of unit cell volumes in the two spaces.

[+++] It is important to note that incoherent and coherent scatterings are not to be confused with direct and interference scatterings. While incoherent scattering does not contain interference effects coherent scattering does include certain direct scattering [see Equation (17)]. The justification for the present convention apparently lies in the explicit display of isotopic effects (and later also spin effects).

The general expression (20) allows excitation of an arbitrary number of oscillators. For the purpose of subsequent discussion we will only consider those inelastic interactions in which one oscillator undergoes a change of energy. The scattering which causes that oscillator to lose or gain an amount of energy equal to n multiples of its level spacing is known as an n-phonon process. Phonon emission or absorption are often used to designate respectively loss or gain of neutron energy. Although all inelastic processes are permissible so long as the energy conservation condition is satisfied, it can be readily shown that the most important single inelastic process is the one-phonon exchange. The one-phonon cross section $\sigma^{\pm 1}$ is obtained from (20) by setting $n_\lambda = \pm\, \delta_{\lambda\lambda_o}$ and using the series representation of $I_n(x)$ appropriate for small arguments.+ Here the oscillator responsible for the scattering is designated by λ_o. The two-phonon cross section $\sigma^{\pm 2}$ is similarly obtained by setting $n_\lambda = \pm\, 2\delta_{\lambda\lambda_o}$, and the ratio $\sigma^{\pm 2}/\sigma^{\pm 1}$ is effectively given by the factor $P_{\lambda_o} e^{-\nu_{\lambda_o}}$ which is proportional to N^{-1}. For this reason it is often sufficient to treat inelastic scattering as essentially a one-phonon interaction. The phonon-absorption process σ^{+n} is related to that for phonon-emission σ^{-n} through the factor $e^{-2n\nu_{\lambda_o}}$. We note that general remarks on energy transfer, made earlier for the Einstein model, apply equally well to the present situation.

+ This is justified since P_λ which appears in the argument contains a N^{-1} factor where N is the total number of particles in the crystal.

While the foregoing discussion applies to both coherent and incoherent scattering it is seen that the coherent cross section contains an additional factor in the form of a delta function. We have already noted that any process must satisfy the energy requirement which for one-oscillator excitation becomes

$$\rho = \pm\ n\,\omega_{\lambda_0}\ . \tag{VII.21}$$

The appearance of the delta function $\delta(\underset{\sim}{\gamma}-2\pi\underset{\sim}{Y})$ imposes an additional condition for coherent scattering. Since $\underset{\sim}{\gamma}$ depends upon the neutron momenta and phonon wave vector this condition may be interpreted as momentum conservation although not in the sense of ordinary particle dynamics. We shall examine in some detail the special case of elastic coherent scattering. For this case the momentum condition is simply

$$\underset{\sim}{\kappa} = 2\pi\underset{\sim}{Y}\ , \tag{VII.22}$$

where $\underset{\sim}{\kappa} = \underset{\sim}{k_f}-\underset{\sim}{k_i}$. For elastic scattering $k_i = k_f$, $\kappa = 2k_i\sin(\theta/2)$ and it is seen that the angle of incidence is equal to the angle of reflection; furthermore, the vector $\underset{\sim}{Y}$ is in the direction perpendicular to the plane of reflection (see Figure 5). For reasons which will be clear shortly let us write

$$\underset{\sim}{Y} = n\underset{\sim}{\tau} = n(u\underset{\sim}{b}^1 + v\underset{\sim}{b}^2 + w\underset{\sim}{b}^3)\ , \tag{VII.23}$$

where we recall that $\underset{\sim}{Y}$ is a vector whose components in the reciprocal lattice are integers. Integer n is such that (uvw) are the smallest integers having the same ratio as the components of $\underset{\sim}{Y}$. Consider now plane A whose intersections with the lattice basic vectors $\underset{\sim}{a}_i\,(i=1,2,3)$ are

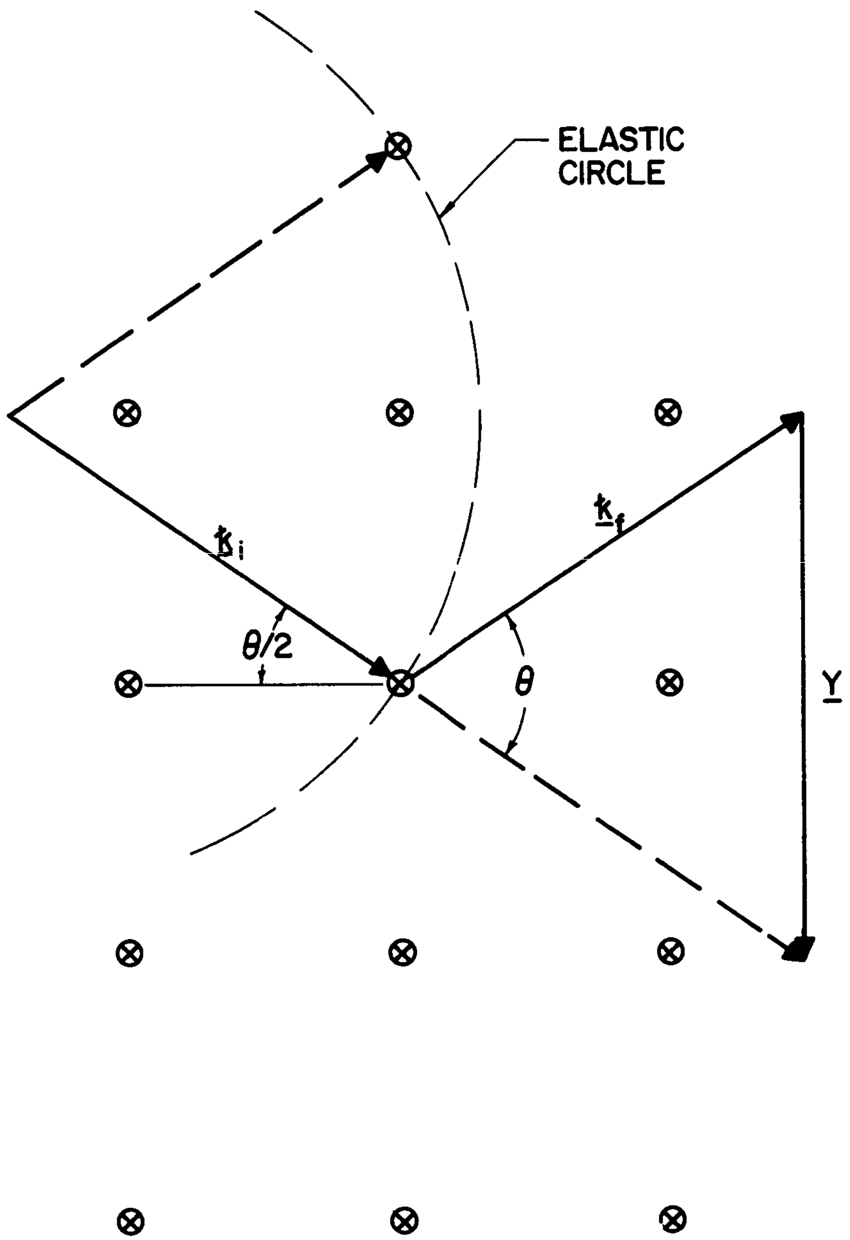

Figure 5. Vector Relation in Reciprocal Lattice Space for Elastic Coherent Scattering.

respectively $1/u$, $1/v$, $1/w$. We observe that the vector $\underset{\sim}{a}_1/u - \underset{\sim}{a}_2/v$ lies in plane A and that the scalar product of this vector with vector $\underset{\sim}{\tau}$ vanishes. Therefore $\underset{\sim}{\tau}$ which is specified by (uvw) is perpendicular to plane A. Moreover the distance between plane A and the next plane which is parallel to it is $\frac{\underset{\sim}{a}_1}{u} \cdot \frac{\underset{\sim}{\tau}}{\tau} = \tau^{-1}$. + In other words, the length of reciprocal vector $\underset{\sim}{\tau}$ is equal to the reciprocal of interplanar spacing of the scattering planes. It is conventional to specify both the reciprocal vector $\underset{\sim}{\tau}$ in reciprocal lattice space and the scattering planes in crystal lattice space by the same set of integers (uvw). These integers are known as Miller indices and are widely used to denote the orientation of a particular crystal plane.

We can now explicitly exhibit the well-known Bragg condition for interference scattering. From (22),

$$4 \sin^2(\theta/2) = (n\lambda\tau)^2$$

or

$$2d \sin(\theta/2) = n\lambda, \quad n=1,2,..., \tag{VII.24}$$

where $\kappa = k(2\pi)^{-1} = \lambda^{-1}$, λ is the de Broglie wavelength of the neutron, and $d = \tau^{-1}$ is the spacing between crystal planes. The factor n is called the order of reflection since its presence allows other wavelengths to scatter from the same plane at the same angle.++ According to the Bragg condition, wich is (24) with $n=1$, the reflection pattern from a given set

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+ We are taking the second parallel plane as one which passes through the origin.

++ We are not interested in the higher order reflections so henceforth we can take n to be unity $(\underset{\sim}{\tau} = \underset{\sim}{Y})$.

of planes will show peaks at certain scattering angles, each peak represents only those neutrons which satisfy the energy-angle relationship.

The condition for elastic coherent scattering, Equation (22), suggests a simple method, often known as Ewald's construction, for determining Bragg scattering. Consider a reciprocal lattice (Figure 5) in which the reduced incident wave vector $\underset{\sim}{k}_i$ is drawn such that it terminates on a reciprocal lattice point. If another reciprocal lattice point lies on a sphere of radius k_i centered at the origin of the incident vector then interference scattering from the planes normal to the reciprocal lattice vector $\underset{\sim}{Y}$ is possible. That is to say, Bragg scattering can occur if the difference in the reduced wave vectors is equal to a reciprocal lattice vector.

A similar construction exists in inelastic coherent scattering where one phonon is excited. The momentum condition becomes

$$\underset{\sim}{k}_i - \underset{\sim}{k}_f \pm \underset{\sim}{q} = \underset{\sim}{\tau} \, .$$

As an illustration we shall consider scattering by Aluminum[11] which has a face-centered cubic structure with four atoms per unit cell, (000); $(\frac{1}{2}\,\frac{1}{2}\,0)$; $(\frac{1}{2}\,0\,\frac{1}{2})$; $(0\,\frac{1}{2}\,\frac{1}{2})$. For Bragg scattering the structure factor is

$$F(uvw) = \sum_{\substack{\text{unit}\\\text{cell}}} e^{i\underset{\sim}{\kappa}\cdot\underset{\sim}{x}_\ell} = \sum_{\substack{\text{unit}\\\text{cell}}} e^{2\pi i\underset{\sim}{Y}\cdot\underset{\sim}{x}_\ell} = 1 + e^{i\pi(u+v)} + e^{i\pi(v+w)} + e^{i\pi(w+u)} \, ,$$

which vanishes unless u, v, w are all even or odd integers. We therefore consider in the reciprocal lattice a plane containing some of these reciprocal lattice points, for example, those for which v and w are the same (Figure 6). For a given incident neutron wavelength the scattering angle

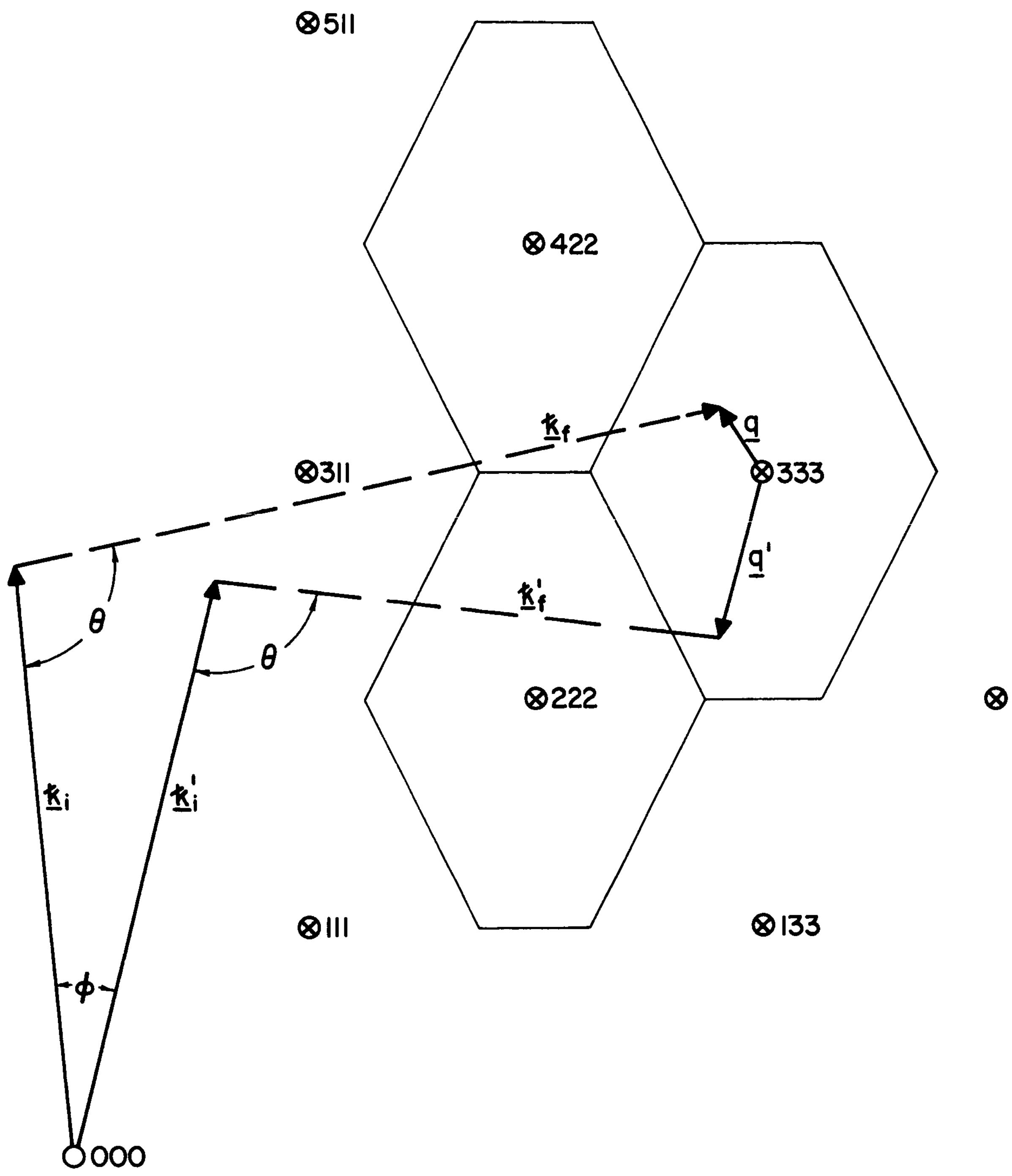

Figure 6. Vector Relations in Reciprocal Lattice for One-Phonon Coherent Scattering by Aluminum.[11] (Not to scale.)

necessary for Bragg reflection by a specific plane is determined from (24) with $n=1$. By knowing this angle, crystal orientations and measuring λ_f, k_f is determined. The phonon vector $\underset{\sim}{q}$ is obtained by connecting $\underset{\sim}{k}_f$ to the nearest reciprocal lattice point. Now suppose the crystal is rotated slightly through an angle φ so that the direction of incoming neutrons is along $\underset{\sim}{k}_i'$. Since the analyzer and scattering angle are fixed a new scattered wave vector $\underset{\sim}{k}_f'$ and a different phonon vector $\underset{\sim}{q}'$ are obtained. Note that the angle Θ is computed for reflection by the 333 plane, but the 511 plane can give rise to Bragg scattering as well.

Thus far we have discussed certain implications of the rather general result given in (20) which is valid for a single crystal whose orientation is specified by the reciprocal vector $\underset{\sim}{Y}$. The differential cross section $\langle\sigma(\rho,\underset{\sim}{\Omega})\rangle$ for a polycrystal, a macroscopic crystal containing many single crystals at random orientation, can be obtained by averaging $\sigma(\rho,\underset{\sim}{\Omega})$ over all crystal orientations or equivalently over all directions of $\underset{\sim}{Y}$, and summing over all the permissible values of $\underset{\sim}{Y}$ which give rise to coherent scattering. If the differential cross section is integrated over all scattering angles and final neutron energies the result will be the total cross section σ for neutron-crystal interaction. Obviously, any attempt to obtain these cross sections for the general case will be a difficult and tedious task. For in addition to the $\underset{\sim}{Y}$-dependence Equation (20) exhibits explicit directional dependence in the vibrational frequencies $\omega_j(\underset{\sim}{k})$ and polarization vectors $\underset{\sim}{A}_j(\underset{\sim}{k})$. At the same time, evaluation of the Debye-Waller factor requires specific knowledge of the dispersion relation.[+] We will, therefore, turn to special cases for subsequent analysis and illustration. Aspects of inelastic scattering will be discussed in terms of one-phonon process while the Debye model is adopted in obtaining the various elastic cross sections. It will be shown that the inelastically scattered neutrons can be used, in certain cases, to determine dispersion relation and vibrational frequency distribution, and that the Debye approximation affords a considerable simplification but still leads to results useful in studying limiting cases.

A. One-Phonon Process and Crystal Dynamics[(12)]

We consider separately the one-phonon differential incoherent and coherent cross sections as obtained from Equation (20),

$$\sigma^{\pm 1}_{\mathrm{incoh}}(\rho,\Omega)_\lambda = \frac{a_I^2 k_f e^{-2W}\hbar\,[\kappa\cdot A_j(q)]^2}{2MNk_i\omega_j(q)}\left\{\left(e^{2\nu_j(q)}-1\right)^{-1} + \frac{1}{2}(1\mp 1)\right\}, \quad \text{(VII.25)}$$

$$\sigma^{\pm 1}_{\mathrm{coh}}(\rho,\Omega)_\lambda = \frac{(2\pi)^3 a_C^2 k_f e^{-2W}\hbar\,[\kappa\cdot A_j(q)]^2}{2MNk_i\omega_j(q)v_a}\,\delta(\kappa\pm q-2\pi Y)\left\{\left(e^{2\nu_j(q)}-1\right)^{-1} + \frac{1}{2}(1\mp 1)\right\}, \quad \text{(VII.26)}$$

where the small argument expression of the modified Bessel function has been used, and upper and lower signs denote phonon absorption and emission respectively.[++] The index λ reminds us that energy is interchanged with the λ-th phonon (previously called λ_0) with propagation vector q and

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[+] Dispersion relation $\omega_j(k)$ is an expression showing the dependence of the circular frequency ω upon the direction of polarization j and wave propagation vector k. This is a fundamental property of the crystal determined by geometrical structure and interatomic forces.

[++] In this notation, $\omega_j(q) = \omega_j(2\pi k)$ is always positive.

polarization j. In other words, energy condition

$$k_f^2 - k_i^2 = \pm \frac{2m_o}{\hbar} \omega_j(\underset{\sim}{q}) \qquad \text{(VII.27)}$$

is satisfied for both cross sections. Equation (26) explicity contains the momentum condition for coherent scattering.

We shall first examine incoherent scattering. According to (27) all final neutron wave vectors $\underset{\sim}{k}_f$ must end inside or on a shell defined by two spheres of radii max. $(0,[k_i^2 - \frac{2m_o}{\hbar}\omega_{max}]^{1/2})$ and $(k_i^2 + \frac{2m_o}{\hbar}\omega_{max})$, where ω_{max} is the maximum value of $\omega_j(\underset{\sim}{q})$ for all $\underset{\sim}{q}$ and j. Since energy conservation is the only condition to be satisfied, incoherent scattering is seen to occur in all directions, and the energy distribution of the scattered neutrons is continuous within the shell. We note that $\sigma^{\pm 1}_{incoh}(\rho,\underset{\sim}{\Omega})_\lambda$ usually is not measured since the experiment cannot isolate the effect caused by the λ-th phonon from that caused by all other phonons. So no information will be lost if we consider incoherent scattering in terms of a cross section that does not depend upon the particular phonon involved in the interaction, i.e., we write

$$\sigma^{\pm 1}_{incoh}(\rho,\underset{\sim}{\Omega}) = \sum_{j,\underset{\sim}{q}} \sigma^{\pm 1}_{incoh}(\rho,\underset{\sim}{\Omega})_\lambda \, \delta(E_f - E_i \mp \hbar\omega_j(\underset{\sim}{q})) \; . \qquad \text{(VII.28)}$$

In most cases N is large so that the summation over $\underset{\sim}{q}$ can be replaced by an integration over the reciprocal unit cell,

$$\sum_{\underset{\sim}{q}}^{N} \longrightarrow \frac{V}{(2\pi)^3} \int_{\substack{\text{unit}\\\text{cell}}} d\underset{\sim}{q} \quad ,$$

where V, the volume of the crystal, is the uniform density of distribution of $\underset{\sim}{q}$ in reciprocal space. The summation over $\underset{\sim}{q}$ in the Debye-Waller factor is similarly treated. Next we eliminate the directional dependence through polarization by restricting the class of crystals under consideration. It is known that for a cubic crystal $\sum_j A_{j\alpha}(q)A_{j\beta}(q) = \delta_{\alpha\beta}$, or

$$\sum_j \int dqG_j(q)A_{j\alpha}(q)A_{j\beta}(q) = \delta_{\alpha\beta}\frac{1}{3}\sum_j \int dqG_j(q) \ .$$

As a result, we get

$$\sigma^{\pm 1}_{incoh}(\rho,\Omega) = \frac{a_I^2 k_f \hbar v_a e^{-2W}}{6(2\pi)^3 M k_i}$$

$$(x) \sum_j \int_{\substack{unit\\cell}} dq \frac{\kappa^2\delta(E_f - E_i \mp \hbar\omega_j(q))}{\omega_j(q)} \left\{(e^{2\nu_j(q)} - 1)^{-1} + \tfrac{1}{2}(1\mp 1)\right\}$$

where

$$2W = \frac{\hbar\kappa^2 v_a}{6(2\pi)^3 M} \sum_j \int_{\substack{unit\\cell}} dq \frac{\coth \nu_j(q)}{\omega_j(q)} \ .$$

The integral can be transformed to an integration over the frequencies by introducing the frequency distribution function $f(\omega)$, defined as the number of normal mode frequencies per unit frequency interval divided by the total number of frequencies. Formally we can write

$$f(\omega)\Delta\omega = \frac{V}{3N}\sum_j (2\pi)^{-3} \int_{\omega \le \omega_j(q) \le \omega + \Delta\omega} dq \ ; \qquad 0 \le \omega \le \omega_{max}$$

where the integration is over a reciprocal unit cell. The incoherent differential cross section then becomes

$$\sigma^{\pm 1}_{incoh}(\rho,\Omega) = \frac{a_I^2 k_f m_o \kappa^2 e^{-2W}}{M|k_f^2 - k_i^2|k_i} f\left(\frac{\hbar}{2m_o}|k_f^2 - k_i^2|\right)\left\{\left(e^{\frac{\hbar^2|k_f^2-k_i^2|}{2m_o k_o T}} - 1\right)^{-1} + \tfrac{1}{2}(1\mp 1)\right\} \ . \tag{VII.29}$$

Equation (29) is independent of the direction of $\underset{\sim}{\kappa}$ and crystal orientation but does depend upon the scattering angle Θ. At a fixed Θ, $\sigma^{\pm 1}_{incoh}$ can be measured as a function of energy transfer and in this manner it is possible to deduce the frequency distribution of the crystal.+

The above treatment cannot be applied to noncubic crystals and therefore incoherent scattering will depend upon crystal orientation. However, as shown by Van Hove,[12,13] certain properties of $f(\omega)$ can still be determined from the energy distribution of scattered neutrons in a given direction. In particular, it is known that $f(\omega)$ for a general crystal contains a finite number of singularities as a consequence of the periodic structure. These singularities are known as singular frequencies and are generally those values where $\underset{\sim}{\nabla}\, \omega_j(\underset{\sim}{q}) = 0$ or equivalently where the discontinuities occur in the first derivative of the energy distribution. For the cubic crystals the energy distribution leads directly to the singularities. Even for noncubic crystals the energy distribution of incoherently scattered neutrons will exhibit similar singularities at energies independent of direction of scattering.

In the case of polycrystals or powder the foregoing results for cubic crystals remain unchanged since (29) is independent of crystal orientation. For noncubic crystals (28) has to be averaged over crystal orientations; however, the singularities can still be observed since their occurrence in the energy distribution is not affected by directional effects.

+ Most dynamical properties of a crystal can be related to its dispersion relation and frequency distribution function. From this point of view the problems of crystal dynamics can be considered as reduced to a determination of these two quantities.

Incoherent scattering due to multi-phonon processes in general gives rise to a distribution dependent upon direction of scattering but is continuously differentiable. All singularities in an observed distribution can therefore be attributed to one-phonon processes.

Now we examine the spectrum of coherently scattered neutrons. As previously noted, for this scattering process there exists an additional condition on the momentum. Combining the two conditions we can write

$$k_f^2 - k_i^2 = \pm \frac{2m_o}{\hbar} \omega_j(\underset{\sim}{\kappa}) \ . \tag{VII.30}$$

In (30) we have made use of the fact that the wave vector $\underset{\sim}{q}$ is defined up to $2\pi\underset{\sim}{\tau}$, where $\underset{\sim}{\tau}$ is any reciprocal vector so that $\omega_j(\underset{\sim}{q}+2\pi\underset{\sim}{\tau}) = \omega_j(\underset{\sim}{q})$. For each j Equation (30) describes a surface in reciprocal space and the three surfaces form the so-called scattering surface S.+ It can be noted that S is continuous and therefore neutrons can be coherently scattered in all directions. On the other hand since the final neutron wave vector must end on the surface the energy distribution along any direction is not continuous (in general, a vector in any direction will cross the surface three or more times). If the discrete neutron energy is measured at a given scattering angle, a point on each layer of S is determined. By repeating this measurement as a function of direction the dispersion relation, $\omega_j(\underset{\sim}{q})$ is thus obtained in terms of the scattering surface. The discrete nature of the energy distribution enables the one-phonon coherent process to be separated from the incoherent and multi-phonon processes. The coherently scattered neutrons in the polycrystal case will exhibit a

+ Properties of S were first noted by Placzek and Van Hove, Reference 12.

continuous distribution on account of the directional average. For this reason, a single crystal rather than powder is more suitable in lattice-vibration experiments.

It is interesting to note that for phonon emission (lower sign) Equation (30) cannot be satisfied if $k_i < k_{min}$, where $k_{min} = \pi\tau_{min}$ and τ_{min} is the smallest reciprocal vector which corresponds to that set of crystal planes with maximum spacing. This is seen to be the Bragg cutoff mentioned in Section II. When $k_{min} < k_i < (\frac{2m_o}{\hbar}\omega_{max})^{1/2}$, the high frequency phonons cannot be excited and certain scattering directions are restricted. It is only when $k_i > (\frac{2m_o}{\hbar}\omega_{max})^{1/2}$ that all phonons can be excited and scattering is allowed in all directions.

The one-phonon coherent cross section is given by (26). Again if we sum over all $\underset{\sim}{q}$ and j subject to energy conservation and replace the $\underset{\sim}{q}$ summation by an integration we obtain

$$\sigma^{\pm 1}_{coh}(\rho,\underset{\sim}{\Omega}) = \frac{\hbar k_f a_C^2 e^{-2W}}{2Mk_i} \sum_j \frac{[\underset{\sim}{\kappa}\cdot\underset{\sim}{A}_j(\underset{\sim}{\kappa})]^2}{\omega_j(\underset{\sim}{\kappa})} \delta(E_f - E_i \mp \hbar\omega_j(\underset{\sim}{\kappa}))$$

$$(x)\ \{(e^{2\nu_j(\underset{\sim}{\kappa})} - 1)^{-1} + \tfrac{1}{2}(1\mp 1)\} ,$$

since both A_j and ω_j are periodic functions of the reciprocal lattice. If we now consider a fixed final neutron energy, $\epsilon_f' \rightarrow E_f' = \frac{(\hbar k_f')^2}{2m_o}$, then

$$\sigma^{\pm 1}_{coh}(\epsilon_f' \rightarrow E_f') = \int dk_f \sigma^{\pm 1}(\rho',\underset{\sim}{\Omega})$$

$$= \frac{\hbar a_C^2[\underset{\sim}{\kappa}'\cdot\underset{\sim}{A}_j(\underset{\sim}{\kappa}')]^2 \left|k_i \pm \left\{\frac{2m_o}{\hbar}\omega_j(\underset{\sim}{\kappa}')\right\}^{1/2}\right| e^{-2W_{\underset{\sim}{\kappa}=\underset{\sim}{\kappa}'}}}{2Mk_i\omega_j(\underset{\sim}{\kappa}')\left|2k_f \mp \frac{2m_o}{\hbar}\left(\frac{d\omega_j(\underset{\sim}{\kappa})}{dk_f}\right)_{\underset{\sim}{k}_f=\underset{\sim}{k}_f'}\right|}$$

$$(x)\ \{(e^{2\nu_j(\underset{\sim}{\kappa}')} - 1)^{-1} + \tfrac{1}{2}(1\mp 1)\} , \qquad \text{(VII.31)}$$

where $\underset{\sim}{\kappa}' = \underset{\sim}{k}_i - \underset{\sim}{k}_f'$ and j is that polarization index for which the energy conservation, $E_f' = E_i \pm \hbar\omega_j(\underset{\sim}{\kappa}')$, is satisfied. The scattered intensity therefore varies as $[\omega_j(\underset{\sim}{\kappa}')]^{-1}$. Since small $\omega_j(\underset{\sim}{\kappa}')$ implies that $\underset{\sim}{\kappa}' - 2\pi\underset{\sim}{\tau}$ is small or $\underset{\sim}{\kappa}'$ is close to a reciprocal lattice point the intensity peaks in the neighborhood of reciprocal lattice vectors. Also for small frequencies the lattice can be considered as essentially isotropic and thus phonon polarizations will be purely longitudinal and purely transverse. The different polarizations can be distinguished from the intensity because $\underset{\sim}{\kappa}' \cdot \underset{\sim}{A}_j(\underset{\sim}{\kappa}')$ is zero and κ' for the transverse and longitudinal branches respectively.

B. The Debye Model and Incoherent Approximation

We obtain from Equation (20) the single crystal, differential elastic incoherent and coherent scattering cross sections,

$$\sigma^o_{incoh}(\underset{\sim}{\Omega}) = a_I^2 e^{-2W} , \tag{VII.32}$$

$$\sigma^o_{coh}(\underset{\sim}{\Omega}) = \frac{(2\pi)^3 a_C^2 e^{-2W}}{v_a} \, \delta(\underset{\sim}{\kappa} - 2\pi\underset{\sim}{\tau}) , \tag{VII.33}$$

where again the small argument expression for the modified Bessel function has been used. As noted previously the polycrystalline cross sections are obtained by averaging (32) and (33) over crystal orientation. This cannot be done without explicit knowledge of the phonon polarization and $\omega_j(\underset{\sim}{q})$. If the crystal is cubic then the previous result applies and the Debye-Waller factor depends only upon scattering angle Θ. In general, however, it is very difficult to solve the dynamical matrix discussed in Section V to obtain $\omega_j(\underset{\sim}{q})$. On the other hand, there exists a useful approximation, originally proposed by Debye for the theory of specific heats, which eliminates the directional dependence in the Debye-Waller factor. The Debye model assumes that the crystal lattice can be treated as an elastic continuum and in this approximation it is shown that the vibrational frequency is independent of direction of polarization and is linearly proportional to the magnitude of the wave vector

$$\omega_j(\underset{\sim}{q}) = c|\underset{\sim}{q}| \;,$$

with c the average sound velocity in the crystal.[+] It is known that this model has been found to give satisfactory results in problems involving a summation of all the vibrational frequencies. Therefore it can be suitably used to predict the energy and temperature dependence of the total cross section.[(4)] But, as we have shown, angular and energy distributions can be sensitive to the details of $\omega_j(\underset{\sim}{q})$ and $f(\omega)$, so in these cases the use of Debye approximation is likely to be inappropriate.

For the purpose of our illustration we shall proceed with the above approximation. The Debye-Waller factor now becomes

$$e^{-2W} = e^{-\frac{\hbar\kappa^2}{2cMN}\sum\limits_{q}^{N} q^{-1}\coth\frac{\hbar cq}{2k_oT}}$$

$$= e^{-\frac{\mu\kappa^2}{M}} \;, \qquad \text{(VII.34)}$$

where

$$\mu = \frac{3\hbar^2}{2(k_o\Theta_D)^3}\int_o^{k_o\Theta_D} x\coth\left(\frac{x}{2k_oT}\right)dx \;.$$

[+] Reader unfamiliar with this aspect should see Reference 8, Chapter II. We note that the Debye model, when applied to a polyatomic crystal, ignores the optical branch of the dispersion relation.

[++] This is not quite true since the longitudinal and transverse velocities are different. For simplicity, the difference is ignored in the present discussion.

In writing (34) the summation of q has been replaced by an integration over the unit cell which could be taken as a sphere with radius $q_{max} = (6\pi^2 N/V)^{1/3}$ chosen such that the total number of wave vectors is N. Θ_D is the Debye characteristic temperature of the crystal defined as $\Theta_D = \frac{\hbar c q_{max}}{k_o}$. Since the Debye-Waller factor is now independent of crystal orientation, $\overset{o}{\sigma}_{incoh}(\underset{\sim}{\Omega})$, as given by (32), is therefore valid for polycrystals as well. The total incoherent elastic cross section is

$$\begin{aligned} \overset{o}{\sigma}_{incoh} &= \int d\Omega \overset{o}{\sigma}_{incoh}(\underset{\sim}{\Omega}) \\ &= \frac{\hbar^2 \pi A a_I^2}{2\mu E_i}\left(1 - e^{-\frac{8\mu E_i}{\hbar^2 A}}\right) , \end{aligned} \tag{VII.35}$$

where $A = \frac{M}{m_o}$. For small E_i, $\overset{o}{\sigma}_{incoh} \longrightarrow 4\pi a_I^2$. The polycrystalline coherent cross section is

$$\begin{aligned} \langle \overset{o}{\sigma}_{coh} \rangle_{\tau} &= \frac{1}{4\pi} \int d\Omega(\underset{\sim}{\tau}) \overset{o}{\sigma}_{coh}(\underset{\sim}{\Omega}) \\ &= \frac{a_C^2}{2 v_a \tau^2} e^{-\frac{\mu(2\pi\tau)^2}{M}} \delta(\kappa - 2\pi\tau) , \end{aligned}$$

where subscript τ denotes coherent scattering from a particular reciprocal vector (or equivalently that set of crystal planes whose normal is parallel to $\underset{\sim}{\tau}$). Since $\kappa = 2k_i \sin(\Theta/2)$, all τ satisfying the inequality $\tau \leq k_i/\pi$ can contribute to coherent scattering.+ Then the total coherent elastic cross section becomes

$$\begin{aligned} \overset{o}{\sigma}_{coh} &= \sum_{\tau \leq k_i/\pi} \int d\Omega \langle \overset{o}{\sigma}_{coh} \rangle_{\tau} \\ &= \frac{\hbar^2 \pi a_C^2}{v_a m_o E_i} \sum_{\tau \leq k_i/\pi} \tau^{-1} e^{-\frac{\mu(2\pi\tau)^2}{M}} . \end{aligned}$$

\+ This implies that at sufficiently low energy where $\tau_{min} > k_i/\pi$ the coherent elastic cross section vanishes. This is another way of stating the Bragg cutoff.

We note that σ^o_{coh} varies as E_i^{-1} and will exhibit jumps whenever a reciprocal vector begins to contribute. At higher energies the effect of additional τ's becomes less and less significant and the fluctuations will eventually diminish. Therefore if we consider sufficiently high incoming neutron energy the summation may be approximated by an integral. Upon multiplying the summand by $4\pi v_a \tau^2 d\tau$ we integrate from zero to k_i/π to obtain

$$\sigma^o_{coh} = \frac{\hbar^2 \pi A a_C^2}{2\mu E_i} (1 - e^{-\frac{8\mu E_i}{\hbar^2 A}}) ,$$

a result which is identical to (35) aside from the factor of scattering length. In this approximation it is then only necessary to calculate the incoherent cross section and replace a_I^2 by $\langle a^2 \rangle$ to obtain the total cross section σ^o. This is known as the incoherent approximation. It is interesting to note that in the low-energy limit the total cross section for a monoisotopic crystal is $\sigma^o = \sigma^o_{incoh} \rightarrow 4\pi\langle a^2\rangle$, often referred to in the literature as the bound-atom cross section.

We will next examine the high-energy limit which corresponds to the interaction between a neutron and a free atom. It is obvious that in this case the neutron energy will be sufficiently high for the incoherent approximation to be applicable. Accordingly, the differential cross section becomes

$$\sigma(\rho,\underset{\sim}{\Omega}) = \frac{k_f\langle a^2\rangle}{2\pi k_i} \int_{-\infty}^{\infty} dte^{-i(E_f-E_i)t} \prod_{\underset{\sim}{k}} e^{-2W} \sum_{n=0}^{\infty} e^{n\nu(\underset{\sim}{k})}$$

$$(x)\ (\frac{P(\underset{\sim}{k})\kappa^2}{2})^n \frac{1}{n!} e^{-in\hbar\omega(\underset{\sim}{k})t} , \qquad \text{(VII.36)}$$

where we are only interested in phonon-emission. The reason is that the above limit implies that we may consider the limit of vanishing temperature; since phonon-absorption is proportional to $\exp\{-\frac{\hbar\omega(\underset{\sim}{k})}{2k_oT}\}$, the probability that the neutron will gain energy therefore vanishes as $T \to 0$. Equation (36) can be rewritten to give

$$\sigma(\rho,\underset{\sim}{\Omega}) = \frac{k_f\langle a^2\rangle}{2\pi k_i}\int_{-\infty}^{\infty} dte^{-i(E_f-E_i)t + \Theta(t)} , \qquad \text{(VII.37)}$$

where

$$\Theta(t) = \frac{(\hbar\kappa)^2}{2MN}\sum_{\underset{\sim}{k}}\left[\frac{\operatorname{csch}\nu(\underset{\sim}{k})}{2\omega(\underset{\sim}{k})} e^{\nu(\underset{\sim}{k})-i\hbar\omega(\underset{\sim}{k})t} - \frac{\coth\nu(\underset{\sim}{k})}{\omega(\underset{\sim}{k})}\right] .$$

As usual we replace the $\underset{\sim}{k}$ summation by an integration in the Debye approximation. At the same time we expand $\exp(-i\omega t)$ in a power series up to order of t^2. Thus

$$\Theta(t) = -\frac{i(\hbar\kappa)^2}{2M}t - \frac{3(\hbar\kappa)^2k_o\Theta_D}{16M}t^2 . \qquad \text{(VII.38)}$$

It is interesting to observe that the expansion requires that $\hbar\omega_{max}t = k_o\Theta_D t < 1$; in other words, $\Theta(t)$ should be large and negative so that for $k_o\Theta_D t \geq 1$ the integrand essentially does not contribute. This will be the case if in (38)

$$\frac{3(\hbar\kappa k_o\Theta_D t)^2}{16M(k_o\Theta_D)} \gg 1, \quad \text{or} \quad \frac{(\hbar\kappa)^2}{2M} = E_R \gg k_o\Theta_D .$$

Therefore the above expansion actually corresponds to the statement that momentum transfer, or recoil energy E_R, is large compared to the binding energy -- the weak binding limit. Before inserting (38) into (37) we note that in order to obtain the free atom result Θ_D must be set equal to zero. This is because our model treats the atom as harmonically bounded in an oscillator potential and so the atom will never become free unless the force constant is identically zero. We therefore obtain from (37)

$$\sigma(\rho,\underset{\sim}{\Omega}) = \frac{k_f\langle a^2\rangle}{k_i}\,\delta(E_f-E_i+E_R). \tag{VII.39}$$

The delta function expresses the physically obvious condition that in the interaction between a neutron and a free atom initially at rest the difference in neutron energies is necessarily the recoil energy of the atom.+ The argument of the delta function can be written as

$$k_f = \frac{k_i}{(m_o+M)}\left\{m_o\cos\Theta + (M^2-m_o^2\sin^2\Theta)^{1/2}\right\} \quad ,$$

or

$$\left(\frac{A-1}{A+1}\right)k_i \leq k_f \leq k_i \quad .$$

Integrating (39) over $\underset{\sim}{\Omega}$, we find

$$\sigma(E_f) = \begin{cases} 4\pi \langle a^2\rangle \dfrac{A}{4E_i} = 4\pi \langle a_f^2\rangle\,[E_i(1-\alpha)]^{-1} \;; & \alpha E_i \leq E_f \leq E_i \\ 0 \;; & \text{otherwise} \end{cases} \tag{VI.40}$$

where $\alpha = \left(\frac{A-1}{A+1}\right)^2$. This particular result, which can be derived from purely kinematic arguments,++ is widely used to describe the neutron energy moderation process in nuclear reactor analysis. Integrating (40) over final energy we obtain $\sigma = 4\pi \langle a_f^2\rangle$, the correct total scattering cross section for a free particle.

+ For a high-energy neutron ($E \gtrsim$ ev) atomic motion can be neglected.

++ It would be necessary to assume that the atom is initially at rest, scattering is elastic and isotropic in the center-of-mass laboratory coordinate system.

VIII. NEUTRON-NUCLEAR SPIN INTERACTION

Thus far our discussion has not considered the presence of spin or intrinsic angular momentum. However, a more complete treatment should include the effects due to this additional degree of freedom. As will be seen in the present section, spin effects constitute another factor of incoherence in the scattering.

Let us re-examine the formulations presented in Section III. Instead of (III.10) the Fermi pseudo-potential describing neutron-nuclear interaction is now cast into a spin-dependent form,

$$V = \frac{2\pi\hbar^2}{m_o} \sum_{\ell}^{N} a(\underset{\sim}{s}, \underset{\sim}{\mathcal{J}}_{\ell})\delta(\underset{\sim}{r}-\underset{\sim}{R}_{\ell}) \ , \tag{VIII.1}$$

where $\underset{\sim}{s}$ and $\underset{\sim}{\mathcal{J}}_{\ell}$ are the intrinsic angular momenta of neutron and the ℓ-th nucleus respectively. In order to exhibit an explicit form of $a(\underset{\sim}{s}, \underset{\sim}{\mathcal{J}}_{\ell})$ recall that in the earlier instance the scattering length a_{ℓ}, which is regarded as an empirical constant to be determined experimentally, was introduced. If we now assert that the neutron-nuclear interaction consists of two parts, only one of which is spin dependent, then we might put

$$a(\underset{\sim}{s}, \underset{\sim}{\mathcal{J}}_{\ell}) = a_{\ell} + b_{\ell}(\underset{\sim}{s}\cdot\underset{\sim}{\mathcal{J}}_{\ell}) \ , \tag{VIII.2}$$

where b_{ℓ} is another empirical constant. This particular form is chosen on the basis of simplicity, conservation of angular momentum (rotational invariance), and inversion (space and time) invariance. Moreover, since $s = 1/2$ higher powers of $\underset{\sim}{s}$ can always be reduced to the linear form.

Having made the adjustment in the potential we proceed accordingly with the following modifications. The system eigenfunction now appears as $\Phi_n X_{m_s}$, where X_{m_s} is a column vector specified by ms, the projection of nuclear spin along the z axis.+ As usual, the set $\{\Phi_n X_{m_s}\}$ is taken to be complete and orthonormal in the space and spin variables of the scattering system. Continuing in a similar manner as before we arrive at an expression for the cross section [see Equation (III.11)]

$$\sigma(\rho,\underset{\sim}{\Omega}) = \frac{k_f}{2\pi N k_i \hbar} \int dt e^{-i\rho t} \sum_{\substack{\ell\ell' \\ s}} \sum_{\substack{m_s m_{s_o} \\ s_o}} P_{m_{s_o},s_o} \langle m_s s|a_{\ell'}+b_{\ell'}(\underset{\sim}{s}\cdot \underset{\sim}{\mathcal{S}}_{\ell'})|m_{s_o}s_o\rangle^{\dagger}$$

$$(x) \langle m_s s|a_\ell+b_\ell(\underset{\sim}{s}\cdot \underset{\sim}{\mathcal{S}}_\ell)|m_{s_o}s_o\rangle \sum_{n_o} P_{n_o} \langle n_o|\Lambda^{\dagger}_{\ell'}(t)\Lambda_\ell|n_o\rangle ,$$

where $|m_s s\rangle$ represents the product of neutron spin function and the set of system spin functions, denoted by s and m_s respectively. $P_{m_{s_o},s_o}$ is the probability that the system and neutron have initial states m_{s_o} and s_o, and other quantities in the equation have meanings previously assigned in Section III. Although it is possible to perform experiments in which neutron and system initial spin states are prepared we will assume that neither the neutron and system spin states can be specified nor can the neutron final spin states be observed (system final states are never observed). Under this condition the cross section, as written above, contain a sum over all spin states.

If we again separate the cross section into two parts $\ell=\ell'$ and $\ell\neq\ell'$, then the spin-dependent factors in the direct scattering and

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+ It is to be noted that the separation of spin dependence from coordinate dependence is not always allowed. It is known that for some polyatomic gases and liquids spin can be coupled to internal degrees of freedom of the molecule. However such a separation is rigorous for most crystals including the monatomic case.

interference scattering are

$$\sum_{m_{s_o} s_o} P_{m_{s_o}, s_o} \langle m_{s_o} s_o | (a_\ell + b_\ell \frac{J_\ell^2 - \mathcal{I}_\ell^2 - s^2}{2})^2 | m_{s_o} s_o \rangle , \qquad \text{(VIII.4)}$$

and

$$\sum_{m_{s_o} s_o} P_{m_{s_o}, s_o} \langle m_{s_o} s_o | (a_{\ell'} + b_{\ell'} \underset{\sim}{s} \cdot \underset{\sim}{\mathcal{I}}_{\ell'})(a_\ell + b_\ell \underset{\sim}{s} \cdot \underset{\sim}{\mathcal{I}}_\ell) | m_{s_o} s_o \rangle , \qquad \text{(VIII.5)}$$

where $\underset{\sim}{J}_\ell = \underset{\sim}{s} + \underset{\sim}{\mathcal{I}}_\ell$. As the present representation does not diagonalize J^2 the indicated matrix elements are not easily obtained. However, it is possible to construct by an appropriate linear combination of the $|m_s s\rangle$ states a representation in which J^2, s^2, and J_z, the component of J along the z axis, are diagonalized. We shall label these states as $|jm\rangle$ such that

$$J^2 |jm\rangle = j(j+1) |jm\rangle$$

$$J_z |jm\rangle = m |jm\rangle ; \qquad -j \leq m \leq j . \qquad \text{(VIII.6)}$$

In this representation, (4) becomes (temporarily suppressing subscript ℓ in the spins)

$$\sum_{jm} P_{jm} \{a_\ell + b_\ell \frac{j(j+1) - I(I+1) - 3/4}{2}\}^2 , \qquad \text{(VIII.7)}$$

where the magnitude of $\mathcal{I}^2$ is $I(I+1)$, and P_{jm} is the probability that initially the scattering system and neutron have total spin j and z-component m. The two values which j can assume are $j = I \pm 1/2$ since the magnitude of the neutron spin is $1/2$. The total number of spin states for the entire system is $2j+1 = 2(2I+1)$, or $(2I+1)$ nuclear orientations each of which corresponds to two neutron orientations. Thus, for (7) we have

$$\sum_{m=-(I+1/2)}^{I+1/2} \frac{m}{2(2I+1)}(a_\ell+b_\ell \frac{I}{2})^2 + \sum_{m=-(I-1/2)}^{I-1/2} \frac{m}{2(2I+1)} (a_\ell+b_\ell \frac{I+1}{2})^2$$

$$= \frac{I+1}{2I+1} a_{\ell_+}^2 + \frac{I}{2I+1} a_{\ell_-}^2 ,$$

where

$$a_{\ell_+} = a_\ell + b_\ell \frac{I}{2}$$

and

$$a_{\ell_-} = a_\ell - b \frac{I+1}{2}$$

are the new scattering lengths corresponding respectively to the cases in which neutron spin is parallel and antiparallel to nuclear spin. Consider next the sum (5) which can be written as

$$a_\ell a_{\ell'} + a_{\ell'} b_\ell \sum_{m_{s_o} s_o} P_{m_{s_o}, s_o} \langle m_{s_o} s_o | \underset{\sim}{s} \cdot \underset{\sim}{\mathcal{J}}_\ell | m_{s_o} s_o \rangle$$

$$+ a_\ell b_{\ell'} \sum_{m_{s_o} s_o} P_{m_{s_o}, s_o} \langle m_{s_o} s_o | \underset{\sim}{s} \cdot \underset{\sim}{\mathcal{J}}_{\ell'} | m_{s_o} s_o \rangle$$

$$+ b_\ell b_{\ell'} \sum_{m_{s_o} s_o} P_{m_{s_o}, s_o} \langle m_{s_o} s_o | (\underset{\sim}{s} \cdot \underset{\sim}{\mathcal{J}}_{\ell'}) (\underset{\sim}{s} \cdot \underset{\sim}{\mathcal{J}}_\ell) | m_{s_o} s_o \rangle . \qquad \text{(VIII.8)}$$

Fortunately the last three terms all vanish because they are of the form

$$\sum_m P_m \langle m | \mathcal{J} | m \rangle ,$$

which corresponds to the average of a vector in a system in which the vectors are randomly oriented. Then (8) becomes simply $a_\ell a_{\ell'}$, or in terms of the new scattering lengths,

$$a_\ell a_{\ell'} = (\frac{I_\ell+1}{2I_\ell+1} a_{\ell_+} + \frac{I_\ell}{2I_\ell+1} a_{\ell_-})(\frac{I_{\ell'}+1}{2I_{\ell'}+1} a_{\ell'_+} + \frac{I_{\ell'}}{2I_{\ell'}+1} a_{\ell'_-}) . \qquad \text{(VIII.9)}$$

Using the above results we can proceed to carry out the summation over ℓ. For the direct scattering term $\ell=\ell'$, we have

$$\sum_{\ell}^{N} \left(\frac{I_\ell+1}{2I_\ell+1} a_{\ell+}^2 + \frac{I_\ell}{2I_\ell+1} a_{\ell-}^2\right) = N\left\{<\frac{I+1}{2I+1} a_+^2> + <\frac{I}{2I+1} a_-^2>\right\} ,$$

and for the interference term, $\ell\neq\ell'$, using (9),

$$\sum_{\ell\ell'}' a_\ell a_{\ell'} e^{i\underset{\sim}{\gamma}\cdot(\underset{\sim}{x}_{\ell'}-\underset{\sim}{x}_\ell)} = <\frac{I+1}{2I+1} a_+ + \frac{I}{2I+1} a_->^2 \sum_{\ell\ell'}' e^{i\underset{\sim}{\gamma}\cdot(\underset{\sim}{x}_{\ell'}-\underset{\sim}{x}_\ell)} ,$$

where isotopic average $< >$ has its usual meaning. After a slight rearrangement the scattering lengths appearing in the incoherent and coherent cross sections are respectively (see VI.20)[+]

$$a_I^2 = \left\{<\frac{I+1}{2I+1} a_+^2> + <\frac{I}{2I+1} a_-^2> - <\frac{I+1}{2I+1} a_+ + \frac{I}{2I+1} a_->^2\right\} ,$$

and

$$a_C^2 = <\frac{I+1}{2I+1} a_+ + \frac{I}{2I+1} a_->^2 . \qquad \text{(VIII.10)}$$

In the monoisotopic case we obtain

$$a_I^2 = \frac{I(I+1)}{(2I+1)^2} (a_+ - a_-)^2 ,$$

and

$$a_C^2 = \left(\frac{I+1}{2I+1} a_+ + \frac{I}{2I+1} a_-\right)^2 .$$

+ A simple example of spin-dependent scattering is the case of scattering by hydrogen. For neutron-proton scattering the ratio of spin-dependent factors for interference versus direct scattering is given by $a_C^2(a_I^2 + a_C^2)^{-1} = .025$ so the interference effects are quite negligible. Conventionally this ratio is often regarded as that of coherent scattering length squared to incoherent scattering length squared, and in this sense scattering by hydrogen is essentially all incoherent. We note that this terminology is somewhat different from that used in this writing. Similar remarks also apply to the case of vanadium.

The above results indicate that incoherent scattering can arise from the existence of either isotopic mixture or nuclear spin. Moreover, from the definition of a_+ and a_- the coherent factor (10) can be written as $\langle a \rangle^2$. Thus it is seen that the spin-dependent part of the interaction potential leads to incoherent scattering whereas coherent scattering depends only upon the spin-independent part of the potential.

APPENDIX A

To prove that $D^{ss'}_{\underset{\sim}{k}\alpha\alpha'}$ is Hermitian observe that

$$D^{ss'}_{\underset{\sim}{k}\alpha\alpha'} = \sum_{\ell} D^{ss'}_{\ell\alpha\alpha'} e^{-2\pi i \underset{\sim}{k}\cdot\underset{\sim}{x}_\ell} ,$$

$$D^{ss'*}_{\underset{\sim}{k}\alpha\alpha'} = \sum_{\ell} D^{ss'}_{\ell\alpha\alpha'} e^{2\pi i \underset{\sim}{k}\cdot\underset{\sim}{x}_\ell} ,$$

where

$$D^{ss'}_{\ell\alpha\alpha'} = (M^s M^{s'})^{-1/2} U^{ss'}_{\ell\alpha\alpha'}$$

is real. Since

$$U^{ss'}_{\ell\ell'\alpha\alpha'} = U^{ss'}_{\ell-\ell'\alpha\alpha'} = U^{s's}_{\ell'-\ell\alpha'\alpha} ,$$

then by writing $\ell'' = \ell-\ell'$ and noting that $\underset{\sim}{x}_\ell = \ell^1\underset{\sim}{a}_1$ we have

$$D^{ss'*}_{\underset{\sim}{k}\alpha\alpha'} = \sum_{\ell''} D^{s's}_{\ell''\alpha'\alpha} e^{-2\pi i \underset{\sim}{k}\cdot\underset{\sim}{x}_{\ell''}} = D^{s's}_{\underset{\sim}{k}\alpha'\alpha} .$$

This proves the Hermiticity of the dynamical matrix. Now it remains to prove Equation (VI.14), $e^{s*}_{\underset{\sim}{k}j\alpha} = e^{s}_{-\underset{\sim}{k}j\alpha}$. Take complex conjugate of (VI.12) to obtain

$$\sum_{s'\alpha'} D^{ss'}_{\underset{\sim}{k}\alpha\alpha'} e^{s'*}_{-\underset{\sim}{k}j\alpha'} = \omega^2_{-\underset{\sim}{k}j} e^{s*}_{-\underset{\sim}{k}j\alpha} .$$

Comparing this equation with (VI.12) we obtain (VI.14) provided, of course,

$$\omega^2_{\underset{\sim}{k}j} = \omega^2_{-\underset{\sim}{k}j} . \qquad \text{(A.1)}$$

To show this, consider the secular equation for the solution of $\omega^2_{\underset{\sim}{k}j}$ in (VI.12)

$$|D^{ss'}_{\underset{\sim}{k}\alpha\alpha'} - \omega^2_{\underset{\sim}{k}j}\delta_{\alpha\alpha'}\delta_{ss'}| = 0.$$

Again by complex conjugation

$$|D^{ss'}_{-\underset{\sim}{k}\alpha\alpha'} - \omega^2_{\underset{\sim}{k}j}\delta_{\alpha\alpha'}\delta_{ss'}| = 0 \ .$$

It is clear that (A-1) follows.

APPENDIX B

In order to prove the operator identity

$$e^A e^B = e^{A+B+\frac{1}{2}[A,B]} , \qquad \text{(B-1)}$$

where A and B commute with the commutator [A,B], consider the differential equation

$$\frac{\partial\psi}{\partial\alpha} = (A+B)\psi ,$$

where operators A, B are independent of α. The solution of this equation is

$$\psi = \psi_o e^{\alpha(A+B)} , \qquad \text{(B-2)}$$

where ψ_o is the value of ψ at $\alpha = 0$. Next consider the transformation

$$\psi = e^{\alpha B}\Phi ,$$

which leads to the differential equation for Φ,

$$\frac{\partial\Phi}{\partial\alpha} = A'\Phi \qquad \text{(B-3)}$$

where

$$A' = e^{-\alpha B} A e^{\alpha B} = A + \alpha[A,B] ,$$

since $[A,[A,B]] = [B,[A,B]] = 0$. The explicit dependence on α enables us to write the solution

$$\Phi = \Phi_o e^{\alpha A + \frac{\alpha^2}{2}[A,B]} ,$$

or

$$\psi = \psi_o e^{\alpha B} e^{\alpha A + \frac{\alpha^2}{2}[A,B]} .$$

Comparison with (B-2) and setting $\alpha = 1$ yield

$$e^{A+B} = e^{B} e^{A} e^{\frac{1}{2}[A,B]} \quad .$$

Interchanging A with B gives the identity (B-1).

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